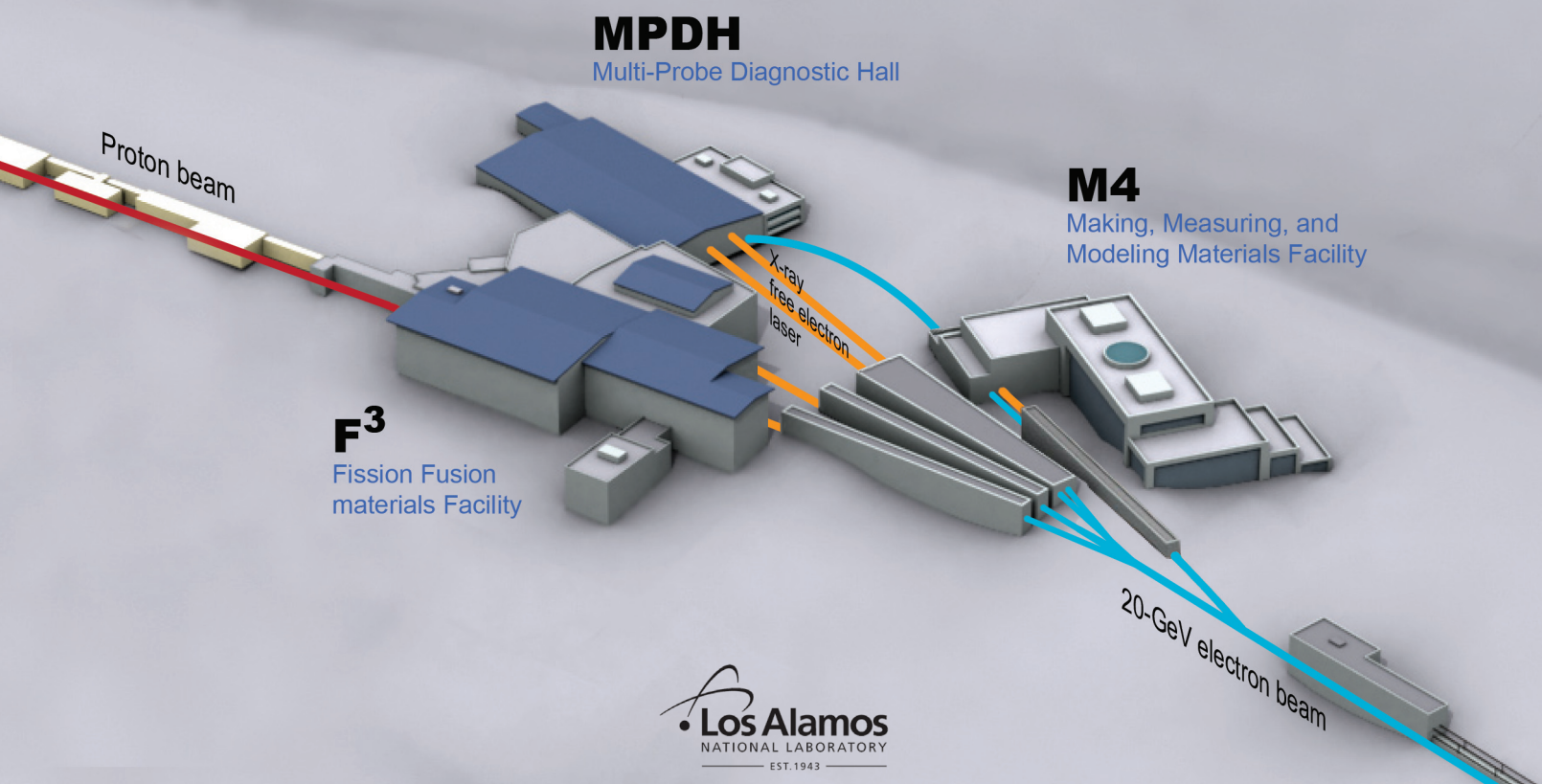


Experimental Physical Sciences

VISTAS

MaRIE: Matter-Radiation Interactions in Extremes



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Susan J. Seestrom
Associate Director
Experimental Physical Sciences

To achieve breakthrough scientific discoveries in the 21st century, an integration of world-leading experimental facilities with capabilities in theory, modeling, and simulation is necessary. In this issue of *Experimental Physical Sciences Vistas*, I am excited to present our plans for Los Alamos National Laboratory's future flagship experimental facility, MaRIE (Matter-Radiation Interactions in Extremes). MaRIE is a facility that will provide transformational understanding of matter in extreme conditions required to reduce or resolve key weapons performance uncertainties, develop the materials needed for advanced energy systems, and transform our ability to create materials by design.

Our unique role in materials science—starting with the Manhattan Project—has positioned us well to develop a contemporary materials strategy pushing the frontiers of controlled functionality—the design and tailoring of a material for the unique demands of a specific application. Controlled functionality requires improvement in understanding the structure and properties of materials in order to synthesize and process materials with unique characteristics.

Today it can take years to develop and qualify new materials for programs—whether it is nuclear weapons or nuclear fuels. Our goal with MaRIE is to accelerate this process by enhancing predictive capability—the ability to compute a priori the material functionality we seek using verified and validated simulation tools. MaRIE will achieve this science-based approach using advanced experimental tools, theoretical models, and multi-physics codes.

Our first issue of *Vistas* focused on our current national user facilities (the Los Alamos Neutron Science Center [LANSCE], the National High Magnetic Field Laboratory-Pulsed Field Facility [NHMFL], and the Center for Integrated Nanotechnologies [CINT]) and the vitality they bring to our Laboratory. These facilities are a magnet for students, postdoctoral researchers, and staff members from all over the world. This, in turn, allows us to continue to develop and maintain our strong staff across the relevant disciplines and conduct world-class discovery science.

The second issue of *Vistas* was devoted entirely to the Laboratory's materials strategy—one of the three strategic science thrusts for the Laboratory. This strategy has helped focus our thinking for MaRIE. We believe there is a bright future in cutting-edge experimental materials research, and that a 21st-century facility with unique capability is necessary to fulfill this goal. The Laboratory has spent the last several years defining MaRIE, and this issue of *Vistas* presents our current vision of that facility.

MaRIE will leverage LANSCE and our other user facilities, as well as our internal and external materials community, for decades to come, giving Los Alamos a unique competitive advantage, advancing materials science for the Laboratory's missions and attracting and recruiting scientists of international stature. MaRIE will give the international materials research community a suite of tools capable of meeting a broad range of outstanding grand challenges.

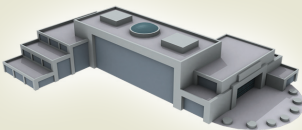
Susan J. Seestrom

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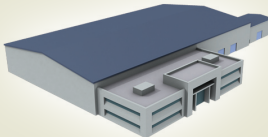
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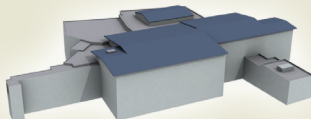
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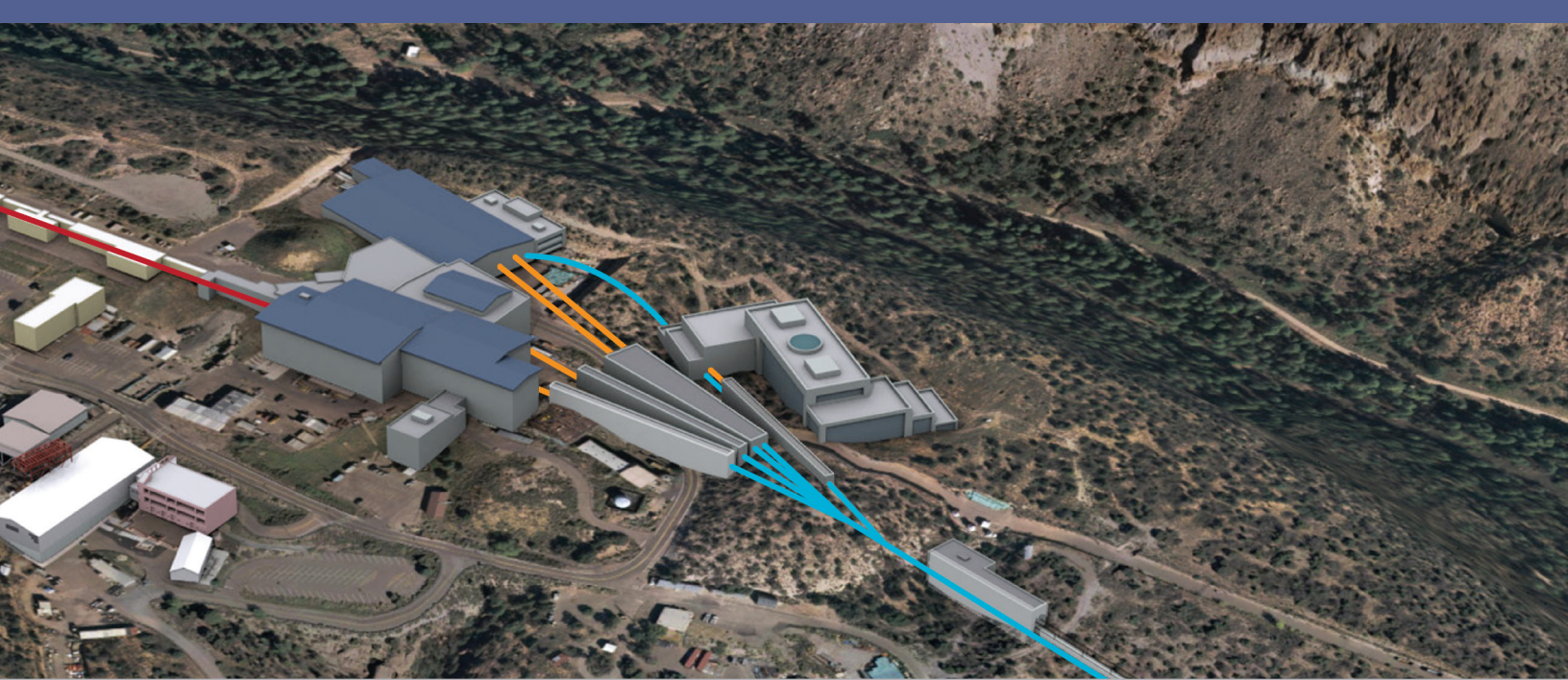


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ON THE COVER:
TOP: The Jemez Mountains viewed from the Rio Grande Valley. Los Alamos, seen in the far left of the image, is located on a series of volcanic alluvial mesas that make up the foothills of the much higher Jemez.

BOTTOM: Concept illustration of the MaRIE facility on the mesa of the Los Alamos Neutron Science Center.



MaRIE: A facility concept revolutionizing materials in extremes

Advanced materials have transformed modern society; for evidence consider the semiconductor advancements that have enabled the creation of handheld devices such as iPods, BlackBerrys, and the other electronic devices that dominate our daily lives. After decades of research, the silicon used in semiconductors operates at or near its intrinsic limits—the result of our understanding the role of atomic-scale impurities and the role of clean-room quality synthesis, factors which previously limited the material's performance.

Here at Los Alamos, as a leader in national security science, we recognize that materials and their associated challenges are ubiquitous, especially the need for materials to perform robustly in extreme environments. The challenge is acute: we know that complex, multi-element materials used for nuclear weapons, nuclear power, and advanced energy systems currently function at one-tenth or less of their intrinsic limits and we lack a predictive understanding of why they fail or how to extend their performance or their lifetime to meet mission requirements.

Great potential exists to change this reality. Materials research is on the brink of a new scientific era in which our traditional approach of observing and validating a material's performance is displaced by our emerging ability to predict and control its function. Progress in this endeavor will stimulate a new generation of national security solutions—from a more reliable nuclear stockpile to higher efficiency clean energy technologies.

Much of today's materials research focuses on nanoscience—the study of matter at the atomic or molecular scale (1 nanometer=1 billionth of a meter or 10^{-9} meters)—because of our desire to exploit the potential in the new phenomena that arise when quantum mechanical effects dominate a material's observed behavior at this scale.

Understanding and utilizing these phenomena in real materials require scientists to conquer the micron frontier, the gap between understanding a material on the atomic scale and validating its performance on a bulk scale. At the micron scale (1 micron=1 millionth of a meter or 10^{-6} meters) defect and grain boundary behavior drives materials strength and damage evolution. The micron frontier is the domain of naturally occurring and artificially induced defects, interfaces, and heterogeneity. The characteristics of a material at the micron scale are what we mean by materials microstructure. Without knowledge of microstructure a material's performance will be limited, but with this knowledge and through directed synthesis we can enhance a material's functionality.

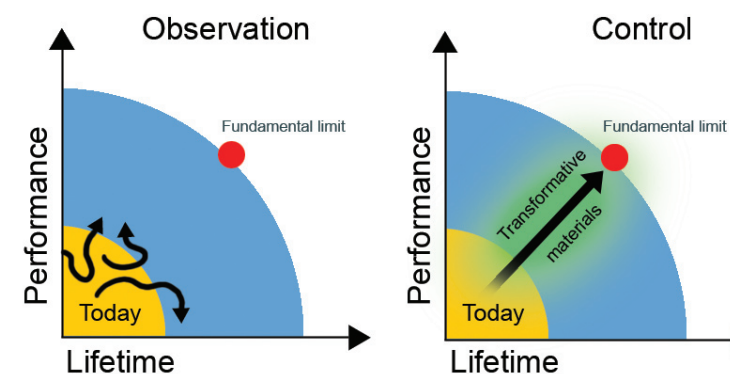
Building on a foundation of materials science leadership

MaRIE, for Matter-Radiation Interactions in Extremes, is Los Alamos's flagship experimental facility intended to revolutionize materials in extremes by conquering the micron frontier and advancing this transition from observation and validation of materials performance to prediction and control of materials functionality.

As discussed throughout this issue of *Vistas*, key to this new era of science is making in situ, transient measurements on real materials in relevant extreme environments using unique simultaneous observational tools.

These tools will be intimately coupled to synthesis efforts in which materials are developed by design through predictive theory rather than discovered by serendipity.

MaRIE builds on many decades of Los Alamos leadership in materials research.



..... Moving from observation to control accelerates materials discovery.

Los Alamos's decision to pursue MaRIE builds on the institution's many decades of materials leadership. In addition to a track record of mission success in actinides and materials dynamics, Los Alamos scientists have been active leaders in many community-based workshops and planning efforts for materials in extremes.

As discussed in previous issues of *Vistas*^{1,2}, materials for the future is one of the Laboratory's three "science that matters" pillars, and the Los Alamos Neutron Science Center (LANSCE), our current signature facility, has been making important contributions in materials research for a number of years.

¹ *Experimental Physical Sciences Vistas: National User Facilities*, Los Alamos National Laboratory, LALP-09-036 (Spring 2009).

² *Experimental Physical Sciences Vistas: Los Alamos Materials Strategy*, Los Alamos National Laboratory, LALP-09-036 (Winter 2010).

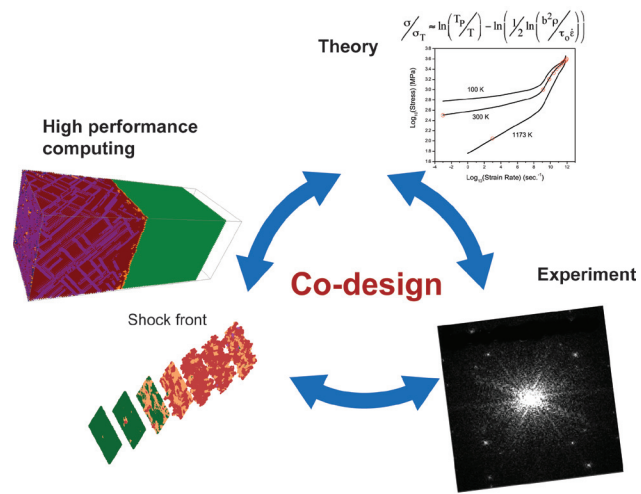
Recent successes by the Laboratory's materials community in peer-reviewed competitions in response to Department of Energy (DOE) solicitations, enabled in part by discretionary strategic Los Alamos research and development investments, indicate that the external scientific community recognizes Los Alamos leadership. These successes include the Consortium for Advanced Simulation of Light Water Reactors and the Center for Materials at Irradiation and Mechanical Extremes.

Now is the time to conquer the micron frontier

Success in conquering the micron frontier requires more than the ability to make measurements on the scale of 10^{-6} meters, a challenge routinely met by a large suite of microscopies and measurement techniques. Success requires simultaneous, in situ, time-resolved measurements of dynamic (rapidly evolving in time) and stochastic (random) processes, especially in extreme environments on the scale of the microstructure. To predict the behavior of complex, multigranular materials undergoing exposure to high radiation or subjected to high rates of strain (for example, structural materials in fission and fusion reactors), researchers need to know both where the atoms are located and where they are not—that is, knowing precisely void size and location inside a three-dimensional, macroscopic solid as a function of time with nanosecond to picosecond resolution.

The advanced experimental tools needed to perform such measurements, including coherent, hard x-ray sources and high-intensity, high-energy proton beams, are just now becoming available. Techniques such as coherent x-ray diffractive imaging and proton microscopy promise a revolution in our ability to predict and control materials functionality. Just as first-generation x-ray free electron lasers (XFELs) are allowing us to watch chemical bonds break and form, these next-generation x-ray sources will catch complex materials in action. Building on the demonstrated success of proton radiography, which was developed at Los Alamos for revealing macroscopic materials dynamic phenomena, proton microscopy is uniquely suited to image micron-scale void formation and evolution within the volumes of dense materials, such as nuclear fuel rod elements in extreme radiation environments.

The availability of advanced diagnostic tools is one reason why now is the time for MaRIE. Advances in multiscale modeling and high performance computing (at the peta- [10^{15}] and exa- [10^{18}] flop scale) are another. For the first time accurate first-principles simulation of microstructure and radiation damage evolution at relevant time and length scales is possible. The ability to validate (or invalidate) these models is a key requirement for predictive understanding of materials functionality.



Validating multiscale models will lead to predictive understanding of materials functionality.

Two examples illustrate the problem that must be solved. When the United States decided to manufacture the plutonium core of a nuclear weapon with a cast process rather than the historical wrought process, it took more than a decade and approximately \$1 billion to certify that the new components would perform as desired. While this is a success story that obviated the need for even larger plutonium production facilities, the country cannot afford such heroic efforts to become the norm. Similarly, today it takes approximately 20 years to certify a new structural material for advanced fission reactors, a pace inconsistent with the needed nuclear renaissance to solve our energy challenge. A process-aware understand-

ing of materials performance is defined as a predictive understanding of how materials processing affects ultimate performance. In both the nuclear weapon and nuclear energy examples, this process-aware understanding is lacking, necessitating a process-based (i.e. identically following the same recipe) specification rather than a product-based (prescribing fundamental materials properties) one. Product-based certification made possible by MaRIE advances at the micron frontier is the faster, cheaper, and better way to a responsive, science-based determination of materials performance.

Advances in diagnostics, modeling, and computing make this the time for MaRIE.

The current state of materials research is a reflection of former Laboratory Director and National Academy of Engineering member Sig Hecker's truism, "Physicists make exquisite measurements on crummy materials and materials scientists make exquisite materials and perform crummy measurements on them." Current national security challenges demand that we perform exquisite measurements on exquisite materials, a microcosm of control science.

Central to this vision of materials-centric national security science leadership is the grand challenge of "control science." Our traditional approach to materials discovery is to "cook and look," in other words, based on intuition, serendipity, or empiricism, we attempt to synthesize a material, measure what we get, and repeat. There is no sense in which "cook and look" should be viewed pejoratively.

Serendipity in the hands of the intuitive has been remarkably successful. However, given a new generation of synthesis tools that allows researchers to manipulate materials at the atomic scale, tools that allow in situ characterization, and theory that can predict properties and performance of materials under extreme operating conditions from structure, it is now possible for us to do better. Multidisciplinary integration coupled closely with usable feedback is essential for success.

The ultimate goal of "materials by design" is the ability to write a chemical formula on a chalkboard and instantly deduce the bulk material properties. While that is a distant goal, it is within our grasp to use theory to direct in which way to guide synthesis or take the next in situ data point in a sample.

A materials research facility for the next decade and beyond

To realize this vision, MaRIE will provide the integrated experimental capabilities needed to achieve transformational materials performance in extreme environments. These integrated tools will be made available to the external scientific community as an international user facility. MaRIE is a facility integrating a state-of-the-art synthesis and characterization capability (M4), a dynamic extremes environment (MPDH), and a materials irradiation environment (F³), with world-class diagnostic tools.

MaRIE's current pre-conceptual definition includes developing a high intensity, low average power XFEL source of 50-100 kiloelectron volt (keV) x-rays, increasing the power of our current 800 megaelectron volt (MeV) proton accelerator to 2 megawatts, constructing measurement halls for in situ multi-probe measurements in dynamic and irradiation extremes, and building an integrated materials synthesis, fabrication, and characterization facility.

In the pages that follow, we describe in more detail the three experimental pillars of MaRIE—their principles, their framework and capabilities, their integration with the entire MaRIE facility, and their advances beyond existing facilities. We also provide examples of experiments already underway that are investigating the discovery science that will be at the heart of MaRIE.

MaRIE achieves transformational materials advances in extreme environments.

Predicting materials performance with MaRIE

The integration of MaRIE experimental data with advanced theory, simulation, and information-theoretic tools will provide an unparalleled capability to understand, predict, and control materials behavior under extreme conditions. The overarching goal is to seamlessly couple theory, experiment, and simulation tools with deliberate feedback in "real time" among all three elements, and use this science-based approach to efficiently predict and control materials properties; we call this integration "co-design."

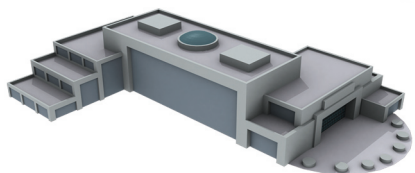
MaRIE enables prediction of material properties from a knowledge of microstructural heterogeneities.

The past half-century has seen enormous advances in our ability to control homogeneous, bulk systems, such as a single crystal of silicon. A number of recent workshops have concluded that we are now at the threshold of a paradigm shift to viewing materials properties as a consequence of intrinsic heterogeneities including defects and interfaces, which we need to characterize and manipulate.³ This new perspective on materials properties requires a highly coupled approach using evolving concepts from theory, simulation, three-dimensional visualization, and experiment to understand and predict microstructure evolution and to design materials with the desired properties. One impact of this endeavor will be the ability to simulate matter in regimes that are not amenable to current experimental techniques and models.

MaRIE will probe the mesoscale, the length scale between a few nanometers and many microns. The mesoscale is the spatial scale where the heterogeneous physics of nucleation, growth, and coalescence takes place. Heterogeneities can occur as a result of spall damage, creation of void and gas bubbles due to fission or irradiation, phase transformations, plasticity, or twinning. It is also the scale where electronic, magnetic, ferroelectric, and photo-induced phases of complex functional materials emerge and coexist.

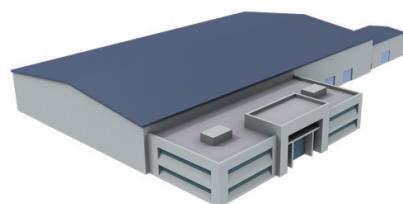
³ Decadal Challenges for Predicting and Controlling Materials Performance in Extremes, Los Alamos National Laboratory, LA-UR 10-02959 (April 2010) and references therein.

M4



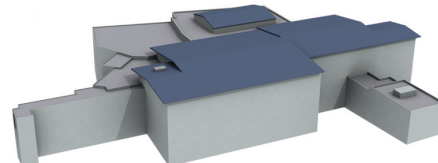
The Making, Measuring, and Modeling Materials Facility (M4) will enable the discovery—by-design of materials to perform with orders-of-magnitude better durability in these extreme environments. Researchers at M4 will also discover the next generation of integrated solid-state solutions for renewable energy and radiation detection. Translating new quantum and nanoscale discoveries to realization for practical applications requires the same capability to bridge the gap from atomic-scale understanding to device performance as that required to understand and exploit the limits of materials strength and irradiation resistance.

MPDH

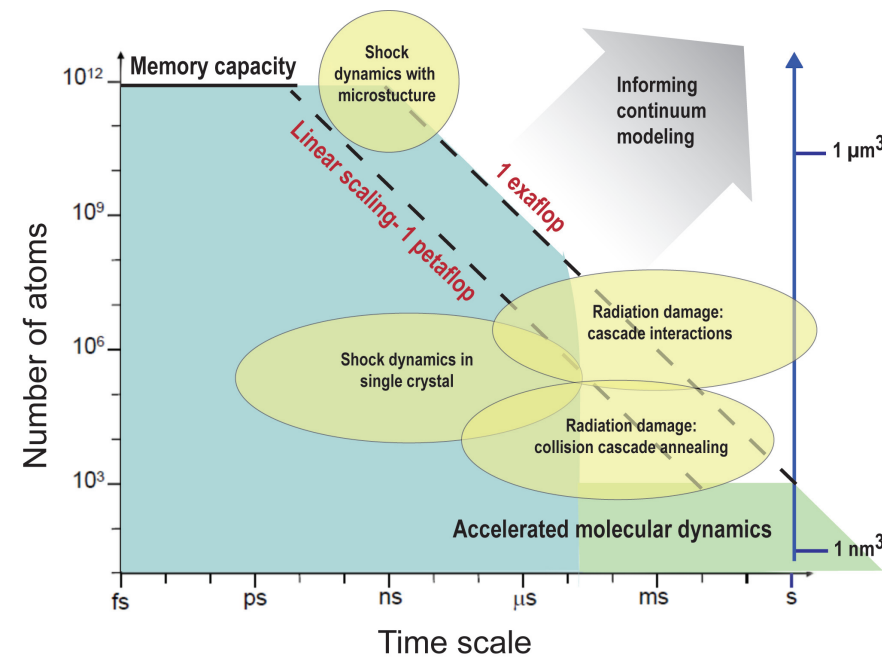


The Multi-Probe Diagnostic Hall (MPDH) will provide unprecedented probes of matter under dynamic extremes including, for the first time, simultaneous x-ray scattering and charged particle imaging measurements of materials interactions at relevant temporal, spatial, and spectral resolutions. These tools will advance the frontiers of dynamic materials behavior spanning solidification phenomena to turbulence in warm dense matter.

F³



The Fission Fusion materials Facility (F³) will provide much needed environments for materials irradiation studies, and even more importantly, will define the frontiers of radiation damage science by advancing the field from post-irradiation materials qualification to in situ diagnostics, thereby providing the ability to better understand and control radiation damage.



- Anticipated advances in petaflop and exaflop computing will enable coarse-scale simulations to spawn sub-scale direct numerical simulations as needed.
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This scale is a challenge to theory, as approximations valid at the extremely small atomistic but fast time scales, or large but slow engineering scales, often break down. In addition, systems driven to extreme conditions are strongly out of equilibrium and can have a widely distributed set of relaxation behaviors. Thus, understanding the physics of the mesoscale is crucial. However, the mesoscale is the least studied of the scales, both experimentally and theoretically, and represents a “hub” through which a perturbation at the macroscale propagates down to smaller scales, and similarly information resident at fine scales emerges and/or couples with other processes to affect properties at higher length scales. This propagation of information across scales is an essential component of MaRIE and emphasizes the need for concurrent measurements, theory, and simulation.

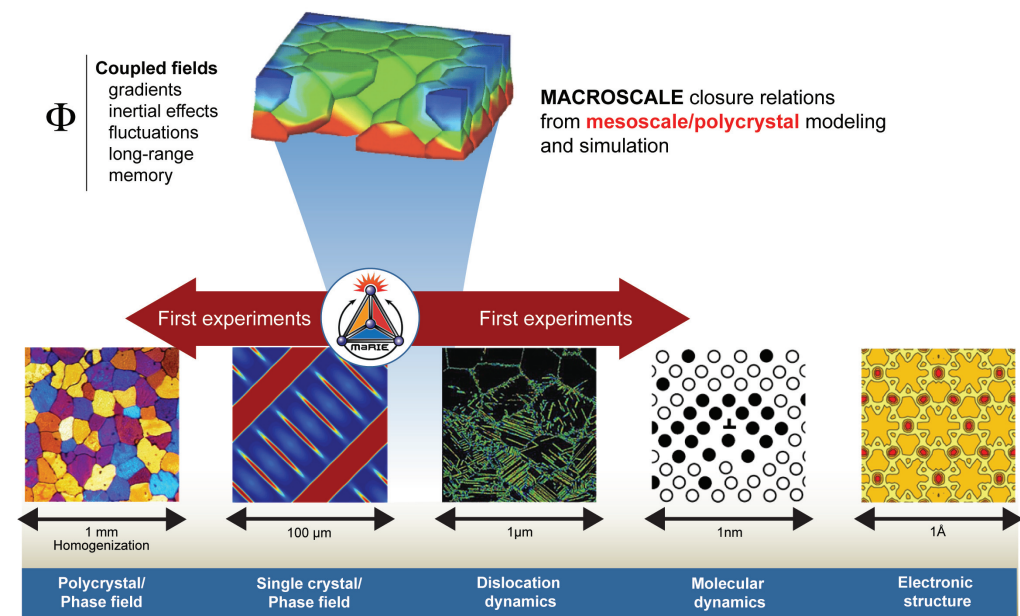
Existing theories and models for functionally heterogeneous processes have largely been developed by making use of the response of homogeneous systems, augmented with simplified averaging schemes. For example, the constitutive equations for materials dynamics are predominantly for ideal phases, or at best assume a “mean-field” description for microstructural effects, such as void damage or phase transformations, and invoke effective rate constants. Currently, data from experiments on recovered samples and microscopic input from atomistic simulations and electronic calculations provide model parameters or other unknowns. The decadal challenge for theory will be to self-consistently incorporate the physics of the various heterogeneous fields into new and highly coupled models. In situ spatial and time-resolved data from MaRIE will help

to calibrate how detailed such models need to be, given that integral engineering-scale applications will demand relative simplicity and ease of calculation. The data will also be used, together with lower length scale predictions, to parameterize the free energies and elastic and inelastic potentials, and to determine transport coefficients for the new models.

Modeling and simulation will be enhanced by the increased computational capacity of extreme (exascale) computing.

The sophistication of modeling and simulation will be enhanced not only by the wealth of data available from MaRIE, but also by the increased computational capacity made possible by the advent of extreme (e.g. exascale) computing.⁴ This will allow novel calculational methods for solving equations, as well as large-scale and time-accelerated atomistic simulations approaching microns in size and milliseconds in time, and electronic structure calculations on unit cells as large as several thousand atoms. Complementing this approach will be inference and sensitivity analyses for estimating statistical fluctuations in microstructural properties, quantifying uncertainties in prediction, tools for the analysis and imaging of scattering and real space data, and the effective use of co-designed high-performance computing resources, all of which will greatly increase resolution and predictive power.

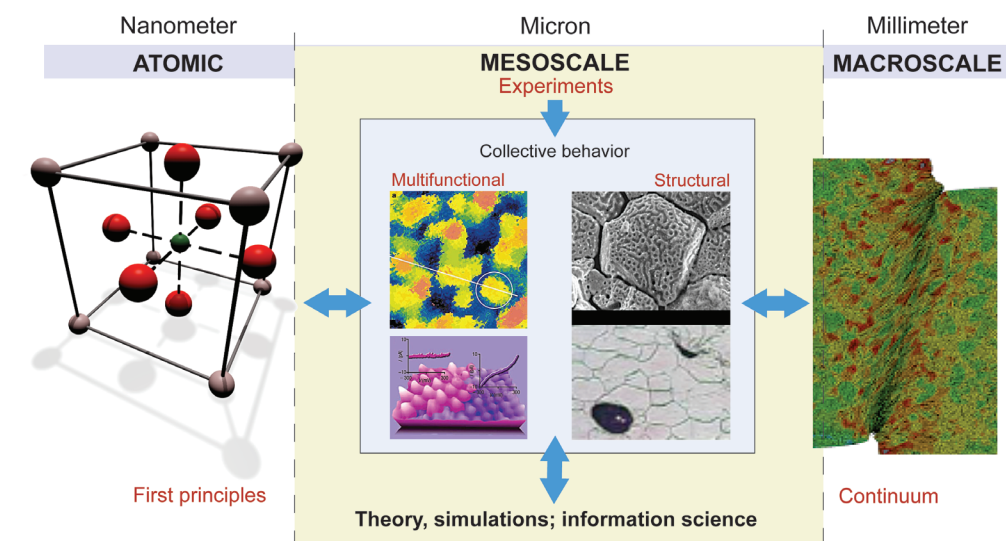
⁴ Scientific Grand Challenges for National Security: The Role of Computing at the Extreme Scale, Department of Energy, PNNL-19379 (2009).



..... Parameterizing and manipulating the free-energy landscape will lead to prediction and control of new materials.

Ultimately, our goal is a coupled, adaptive strategy in which methods at certain scales modify, and are in turn modified by, those at other scales. The process, if carried out self-consistently and combined with uncertainty analysis, provides a road map towards materials prediction with quantifiable variabilities and uncertainties.

A framework to describe the competing states of matter that MaRIE will probe is provided by considering them to result from a “rugged free-energy landscape” that depends on the various order parameter fields associated with the heterogeneities. Consequently, prediction and control will follow from manipulating this energy landscape with external fields such as chemical concentration, stress, and electric, optical or magnetic fields, to optimize a given state for a desired response or functionality. Characterizing and parameterizing such landscapes and exploring the associated nonequilibrium and nonadiabatic behavior are at the heart of MaRIE. For example, through MaRIE we will control the strain rate at which dislocation plasticity or twinning deformation mode is preferred, exploit interfacial properties of alloys that optimize radiation tolerance, and design entirely new classes of materials, such as lead-free piezoelectrics, by effecting and exploiting small energy barriers to yield large polarization.



MaRIE will seamlessly couple theory, experiment, and simulation through ‘real-time’ feedback.

- Lying between atomic scale and macroscale, the mesoscale is crucial, both experimentally and theoretically for advancing materials research.
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The importance of materials for nuclear weapons

President Obama has made it clear that a safe, secure, and effective (and as small as possible) nuclear deterrent is an essential component of United States national security as outlined in the Administration's Nuclear Posture Review. There is also a growing national consensus that investment in sustaining science and technology capability is essential in effectively stewarding a smaller stockpile. The FY11 budget request offers a positive first step in providing the fiscal resources needed by Los Alamos to sustain the nuclear deterrent into the future.

Recently, Los Alamos National Laboratory has made significant strides in advancing science and technology capability in support of Stockpile Stewardship—with the successful execution of several hydrotests utilizing both axes of the Dual-Axis Radiographic Hydrodynamic Test (DARHT) facility and with our participation in many experiments in preparation for ignition at the National Ignition Facility (NIF). In addition, our proton radiography capability at LANSCE continues to inform our understanding, particularly in the area of high explosive burn. We are also now defining the set of first experiments that will be fielded at the Advanced Photon Source on the proposed Dynamic Compression–Collaborative Access Team (DC-CAT) beamline. Finally, we have started on the path towards process-aware understanding, by designing and conducting experiments that test material properties (e.g., strength) under dynamic loading conditions and evaluating the response within the context of the microstructural changes that the processing of the material has induced.

The advent of petascale computing on Roadrunner is providing materials simulations with previously unimaginable resolution. The march towards exascale computation is also actively underway. Los Alamos National Laboratory and National Nuclear Security Administration leadership in experimentally validated simulation science has been broadly recognized by the scientific community and is central to recent efforts to define exascale co-design centers and to advance the frontiers of computational materials design.

MaRIE exemplifies the types of capabilities needed to ensure that Los Alamos retains this leadership in key elements of weapons science. MaRIE will revolutionize materials in extremes, a challenge that spans a broad range of Department of Energy missions, including energy and weapons programs, through unprecedented



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in situ, transient measurements of real materials in relevant extremes, especially dynamic loading and irradiation extremes. There are at least three areas in which MaRIE capabilities can address specific weapons needs:

1. Sustainment of the current stockpile—developing much better predictive capabilities for materials lifetime and failures.
2. Rebuild and life extension—developing the capability to provide materials “by design” to overcome costly “re-learning” of old processes and attempts to duplicate old materials.
3. Weapon performance—developing an understanding of the relationship between material microstructure through material performance to nuclear performance.

Quite recently, Department of Energy Under Secretaries D’Agostino, Johnson, and Koonin acknowledged the importance of LANSCE to Stockpile Stewardship over the next decade and asked Los Alamos National Laboratory Director Anastasio to provide a plan regarding the full suite of issues that need to be addressed in order to sustain LANSCE’s current level of operations through the decade. They also asked the Laboratory to address the longer-term need for high-quality experimental science capabilities and suggested materials under extreme conditions as a potential focus area. In the Laboratory’s response, Director Anastasio asserted that “MaRIE ... is the appropriate signature experimental facility for LANL” and that “we believe that a mission need for a materials-in-extreme facility focused on in situ transient measurements exists.”

This assertion was based in part on a series of five workshops sponsored by the Laboratory in 2009 and culminating in December 2009’s “Decadal Challenges for Predicting and Controlling Materials Performance in Extremes.” These workshops engaged more than 225 scientists from 80 institutions in identifying scientific challenges and research directions to achieve predictive materials performance in extreme environments, focusing specifically on needed capabilities and tools.

MaRIE’s External Advisory Board recently concluded that “*It is particularly noteworthy that these workshops were not LANL-centric, i.e. were not focused on the use of MaRIE as a tool, but rather were focused on defining technical challenges in broad areas of research, identifying needs.*” In fact, many of the scientific challenges are directly relevant to our mission, including understanding the mechanisms of failure under dynamic loading conditions. For example, a predictive understanding of microstructure-based heterogeneity and its consequences for materials damage and failure and phase transformation initiation is presently lacking, but needed for “process-aware” understanding and to evaluate the impact of processing on performance. To understand these impacts, we will use MaRIE’s proposed XFEL and proton radiography simultaneously to probe well-characterized samples that are shock or ramp loaded, and dynamic x-ray diffraction or ankylography to track defect generation and annihilation, with the data gathered used to validate atomistic to mesoscale models of the dynamic response.

Based on these recent positive outcomes, the current focus of MaRIE activity is on translating the identified science need to a specific facility definition and working with DOE, including the Under Secretaries, to define a sustainable acquisition strategy that spans the entire Department. The past year has seen much activity focused on science and technology capabilities required to steward the stockpile, both nationally and locally. Accelerating these activities holds great promise for sustaining important Laboratory capabilities.



B-2 Spirit refueling over the Pacific near Guam. Included in the B-2’s diverse payload capability is the B-61 tactical and strategic nuclear weapon, developed at Los Alamos.

Making, Measuring, and Modeling Materials Facility: Synthesizing and characterizing materials with MaRIE

MaRIE's Making, Measuring, and Modeling Materials (M4) Facility aims to accelerate the transition from observation to control of materials by providing unique synthesis and characterization tools to advance the frontiers of materials design and discovery.

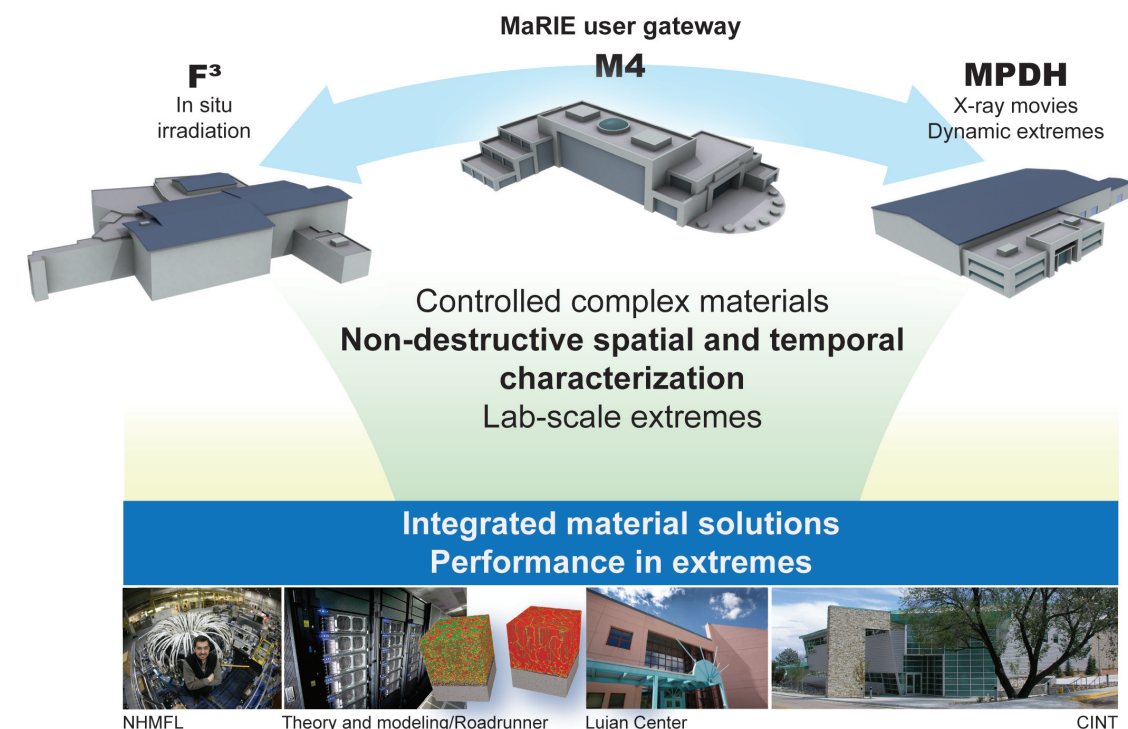
Integration is a key element in the success of M4 and MaRIE as a whole that will enable success in the following:

- Materials by design through the integration of modeling, making, and measuring with rapid feedback
- Controlled synthesis of complex materials through the integration of making and measuring to provide in situ structural information during synthesis
- Improved material lifetimes via a mechanistic understanding of failure through the integration of extremes and time-resolved characterization techniques

To achieve this integration will require effective collaboration among M4 researchers and external users and minimizing the cycle time between making, measuring, and modeling. Bringing together experimentalists, theorists, modelers, and computational scientists in one location fosters spontaneous discussions and ideas and accelerates the theory–synthesis–characterization feedback loop by overcoming genuine issues of sample transport, including moving actinides or explosives and maintaining the integrity of the sample environment.

Advanced facilities for microelectronics such as Sandia National Laboratories' MESA (Microsystems and Engineering Sciences Applications) and the College of Nanoscale Science and Engineering at

With knowledge of microstructure (background image) and through directed synthesis, we can enhance a material's functionality using MaRIE.



..... Researchers entering through the MaRIE user gateway will also access the Laboratory's broader suite of user facilities.

M4 has unsurpassed capabilities to accelerate the discovery of high performance materials for extreme environments.

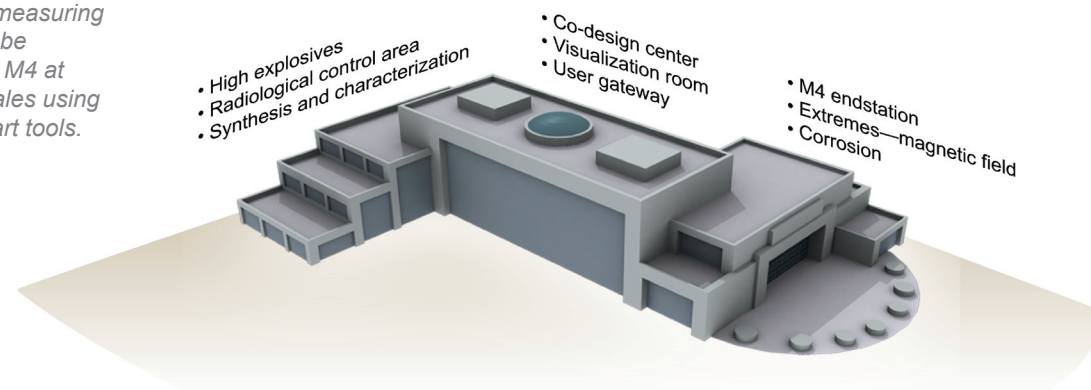
the University at Albany, State University of New York, demonstrate how success is linked to a facility with integrated synthesis and in situ characterization. The Nanoscale Science Research Centers sponsored by the DOE's Office of Basic Energy Sciences, including CINT, emphasize the importance of bringing theory, synthesis, and characterization together under one roof. To provide the same capability for materials in extremes, we must go far beyond the relatively small set of materials used in the microelectronics industry and explore complex multifunctional materials. We must also go beyond the nanoscale and tackle the difficult challenge of micron-scale inhomogeneities that can control both performance and failure. M4 will effectively integrate multiple synthesis techniques and provide in situ characterization in a highly controlled environment to accelerate the discovery of a diverse set of solid-state composite materials designed to perform better and longer in extreme environments. To be successful, M4 will leverage complementary capabilities at Los Alamos, including CINT for nanoscale systems, supercomputing capabilities for micron-scale modeling, the Sigma Facility for large-scale processing, the Plutonium Facility for significant quantities of actinides, the NHMFL for magnetic field extremes, and the Lujan Neutron Scattering Center for neutron-based characterization techniques.

In addition to serving as an integrated synthesis and characterization capability, M4 will serve as a gateway to MaRIE as well as Los Alamos's broader suite of user facilities. At the center of M4 will be a co-design capability dedicated to uniting theory efforts and experiments across length scales. A visualization hub, a locus for the simultaneous examination of computational results and experimental data, will provide users with an ideal opportunity to interpret and compare complex data with the results of predictive simulations. Within the characterization labs, deployed visualization stations will enable real-time assessment of synthesis and processing methods and direct parameter changes to increase control of materials samples.

Integrated synthesis across scales

To achieve the goal of controlled functionality, the ability to integrate synthetic approaches across scales—atomic to meso to macro—will be essential. At the **atomic- and molecular-level scale**, chemical and biological synthesis facilities will provide researchers the ability to make atomic-scale surface modifications critical to corrosion and adhesion; create advanced polymers and electrolytes designed to perform in chemical and temperature extremes; prepare precursor solutions for chemical deposition techniques; assemble bio-inspired templated architectures; and generate large-scale quantities of well-defined nanostructures.

..... Making and measuring activities will be conducted in M4 at all spatial scales using state-of-the-art tools.



At the **mesoscale**, thin-film growth techniques will coalesce in a common experimental chamber allowing multiple methods to be used on a single sample within a controlled environment. These techniques include atomic layer deposition, molecular beam epitaxy, energetic neutral atom beam lithography and epitaxy, and metal organic chemical vapor deposition. Complementing the thin-film growth facility, a fabrication facility with combined deposition and lithography techniques will provide two-dimensional control for meta-material growth, for the fabrication of in situ transmission electron microscopy chambers, and for the fabrication of patterned surfaces. These patterned surfaces can provide position indexing, seed templating for architectural growth, and electrical connections for device testing.

Single crystal growth laboratories will provide high-quality, well-defined bulk samples of metals oxides, nitrides, and carbides and will support the growth of actinide crystals, high explosives, and traditional materials in extreme pressures, thus allowing researchers to create metastable states. “Historically, those institutions that develop new materials are the ones with the best chance to exploit the resulting science and technology opportunities.”⁵ As an example, the impact of crystal growth advances developed at Bell Laboratories on micro-electronics and telecommunications cannot be overstated. Single crystals that can be precisely characterized and modeled will continue as the benchmark for defining new material properties. M4 single crystal growth will be integrated with thin-film deposition, nanostructures, and fabrication techniques to provide bulk single crystals, high-quality interfaces, and crystalline composite materials.

At the **bulk scale**, the microstructure processing facility will control microstructure during casting, welding, powder synthesis, and thermomechanical processing using in situ characterization to observe and ultimately control grain growth and phase formation.

M4 will enable integration across scales to achieve controlled functionality in materials.

⁵ Committee for an Assessment of and Outlook for New Materials Synthesis and Crystal Growth; National Research Council, *Frontiers in Crystalline Matter: From Discovery to Technology* (National Academies Press, 2009).

In situ structural and functional characterization

A suite of intermediate-scale tools will be housed at M4 allowing characterization of chemistry, phase, and structure from atomic to micron scales to bulk sample volumes, and functional/physical property measurements. Structural and functional characterization capabilities will include electron microscopy, scanning probe microscopy, and spectroscopy as well as probes of materials functionality, including electronic and chemical properties and performance figures of merit of integrated devices. Close integration of microscopy techniques will provide micron field-of-view images coupled to sample preparation for nanoscale information relating composition and structure to function. A scanning probe microscopy facility will allow researchers to image surfaces with nanoscale resolution, enabling the correlation of electronic and lattice structure and morphology with material functionality through the measurement of a host of properties on the atomic scale to nanoscale. This microscopy facility will specifically facilitate measurement on the atomic scale to nanoscale of those properties critical to designing materials optimized for performance in extremes, including chemical structure; charge and surface potential distribution; porosity; defect centers; density of states; mechanical response; transport; and magnetic state. A spectroscopy facility will include optical characterization tools to allow study of structure and dynamics of materials on energy scales ranging from 10^{-5} – 10^3 electron volt (eV) and on time scales from 10^{-12} seconds to static. An ultrafast laboratory will explore electron, lattice, and spin degrees of freedom, and energy transfer and relaxation processes in complex materials on their relevant time scales, which range from femtoseconds to nanoseconds.

In all of the structural and functional property measurement suites the emphasis will be placed on advanced non-destructive in situ measurements. These in situ techniques will be used to provide controlled synthesis at different scales, such as optical spectroscopy to observe surface interfaces and particle formation; transmission electron microscopy to observe solidification and crystallization events as they occur; and x-ray diffraction and tomography of bulk samples with specialized environmental cells to probe structure formation during synthesis. In situ microscopy techniques will also be used to

provide insight into how materials evolve and fail in extremes. This will require micron-scale dynamic observations in extremes. The extremes will include radiation extremes through an ion beam laboratory designed to provide high dose rates with small penetration depth and dynamic extremes using laboratory-scale laser drivers. The facilities at M4 will provide rapid throughput and complement the extreme environments at F³ and MPDH. One of the most challenging tasks is to observe the detailed structure at a buried interface as it is formed and as it evolves in extremes. To tackle this problem an endstation driven by MaRIE’s hard x-ray XFEL will be located in M4. This endstation will allow dynamic characterization both during synthesis and in a combination of environments ranging from high electric or magnetic fields, to corrosive environments and high static pressures.

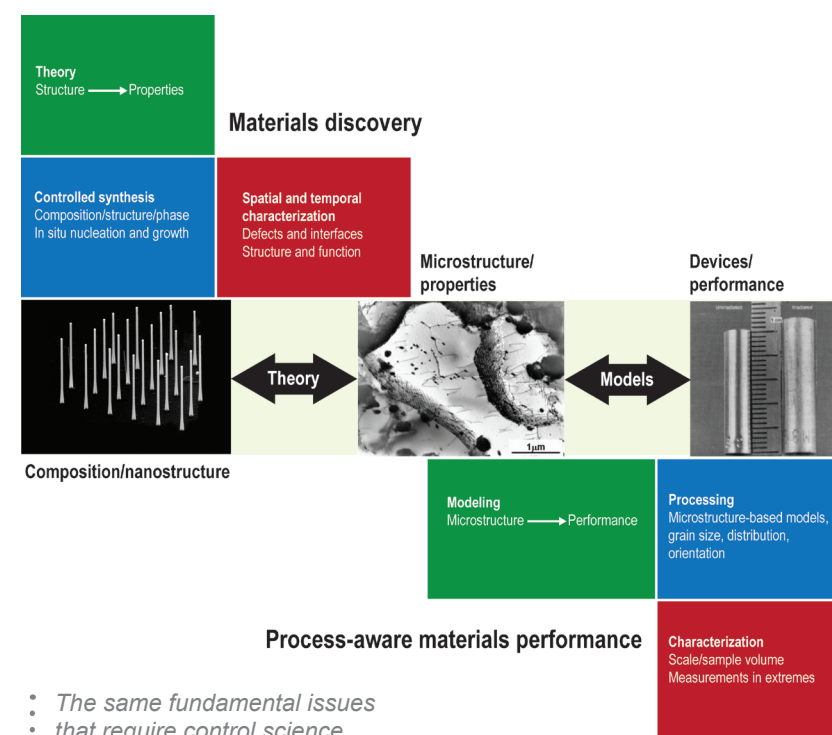
M4 and the transition from observation to control

To develop the advanced materials of the future in a cost-effective and efficient manner, a fundamental shift in the approach to design is required. Traditional research focuses on starting from a particular atomic composition and observing materials properties or measuring a sample microstructure and observing performance. To predict properties in this manner, all (or at least many) potential combinations and

architectures must be explored, with the immediate realization that there are simply too many possibilities and combinations to investigate in a realistic time frame. To succeed in materials design, we must reverse this process by challenging theory and modeling principles—starting with a material’s desired function and working backwards to predict compositions and microstructures resulting in the desired performance. This conceptual change requires a fundamental scientific understanding to develop and validate theories and models. In this mode of research and development, a co-design approach is used to determine the figure of merit optimizing a specific functionality. Exploring this figure of merit over a class of materials in a closely coupled theory-simulation-synthesis-characterization effort will result in a set of design rules to direct materials discovery with desired properties.

M4 is a design center for advanced materials, connecting controlled synthesis and time-resolved characterization in extremes with predictive modeling.

Advanced drug discovery, in which there is only one relevant atomic length scale and the system is treated as a homogeneous solution, is an example of effectively outlining a material’s performance requirements and developing its structural reality. Theory and modeling can accurately predict protein-drug interactions and direct drug design once the protein structure has been well characterized.



• The same fundamental issues
• that require control science
• for materials discovery also
• address process-aware materials
• performance.

Complex materials in extreme environments exhibit tremendous additional degrees of freedom in the form of defects and interfaces at the mesoscale, which are critical to a material’s properties and performance. Acceleration of materials discovery requires a connection between atomic-scale structure and bulk performance under extreme conditions. Today, we predominantly rely on two measurements—one prior to the extreme exposure and one after the extreme exposure. Theories then try to predict performance based on unobserved kinetic processes. To push the forefront of materials by design, we must reveal these mesoscale kinetic processes through in situ transient measurements. Similarly, instead of a single observation on a complex composite, such as radiation-resistant oxide dispersed steel, a nuclear fuel rod, or high explosive, we must characterize materials at different length and time scales on multiple samples having controlled variation in the microstructure in order to ultimately yield predictive design principles. This challenge requires integrating theory and experiment to design materials, coupling synthesis and processing techniques with characterization tools

Nanoscale integration:

A first step towards MaRIE

Integration of nanoscale materials can enable new materials properties and new performance not currently possible in bulk materials. Integration involves assembling diverse nanoscale materials across length scales to design and achieve new properties and functionality.⁶

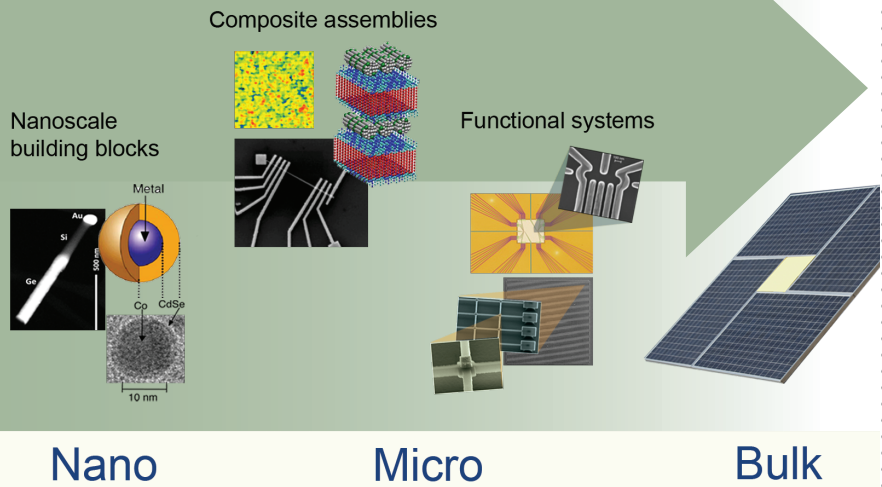
In the past, integration has provided an essential step for the advance of new technologies, from the well-known integrated circuit for computer chips to the DNA microarrays for genetic studies in biomedicine. Combining structures to form heterogeneous nanomaterials is expected to play an equally important role, not only in future materials and device technologies, but also in scientific discovery of materials that may have new properties.

CINT at Los Alamos and Sandia National Laboratories is an important resource en route to MaRIE for the discovery of integrated nanomaterials and provides an environment where functionality is optimized for performance in extreme environments.

Such multifunctional materials based on nanoscale integration will offer a possible new route to design materials with exceptional performance under extreme conditions for MaRIE investigations. The next advances in multifunctional materials will be incorporating nanomaterials into complex composite structures. MaRIE will provide the means to design, synthesize, and characterize the complex, buried interfaces of micron-scale composites in extremes where unique emergent properties can provide exceptional performance.

6 S.T. Picraux, *MRS Bulletin* 35 (4) (2010)

Assembly at the nanoscale leads to new properties and functionality at the macroscale.



Length scale

to provide in situ control of defects and interfaces as they are formed, and coupling multiple characterization tools spanning length scales and sample volumes to watch the defects and interfaces evolve in extremes.

The same fundamental issues requiring control science for materials discovery also enable process-aware materials performance. In this case, we need to again shift the paradigm from one of modeling with empirical input parameters optimized to fit the data without significant scientific basis to one where the theoretical codes include fundamental materials and microstructure properties controlled by processing conditions. The current “product design and development cycle now outpaces the materials development cycle.”⁷ M4 will help reverse this trend by fostering the concept of integrated computational materials engineering in which “materials information, captured in computational tools is integrated with engineering product performance analysis and manufacturing-process simulation.”⁸

M4 as a pillar of an integrated MaRIE facility

M4 will allow controlled synthesis of complex materials, provide nondestructive characterization, and create lab-scale extremes designed to complement those at F³ and MPDH. New radiation and dynamic shock extremes will be provided in F³ and MPDH for exploring centimeter-scale samples, but complementary extremes in the form of a multi-ion beam facility and small-scale laser-induced dynamic extremes will be housed in M4. These will provide accessible, high-throughput capabilities. The ion beam facility will allow for high-dose levels on a large number of small samples not possible at F³.

7 Committee on Integrated Computational Materials Engineering, National Research Council, *Integrated Computational Materials Engineering: A Transformational Discipline for Improved Competitiveness and National Security* (The National Academies Press, 2008).

8 *Decadal Challenges for Predicting and Controlling Materials Performance in Extremes*, Los Alamos National Laboratory, LA-UR 10-02959 (April 2010).

MaRIE SCIENCE: Process-aware materials

Predicting performance through knowledge of the fabrication process

Process-aware materials performance is a materials design approach, which for a given set of performance requirements, utilizes predictive modeling to select the necessary composition, microstructure, and processing pathways for materials synthesis. This approach enables researchers to efficiently explore a large number of processing variables, thereby allowing targeted experiments to accelerate research, development, and certification in nuclear reactor applications.

An expert’s take on materials design and discovery



George Crabtree
Senior Scientist and
Distinguished Fellow
Materials Science Division
Argonne National
Laboratory

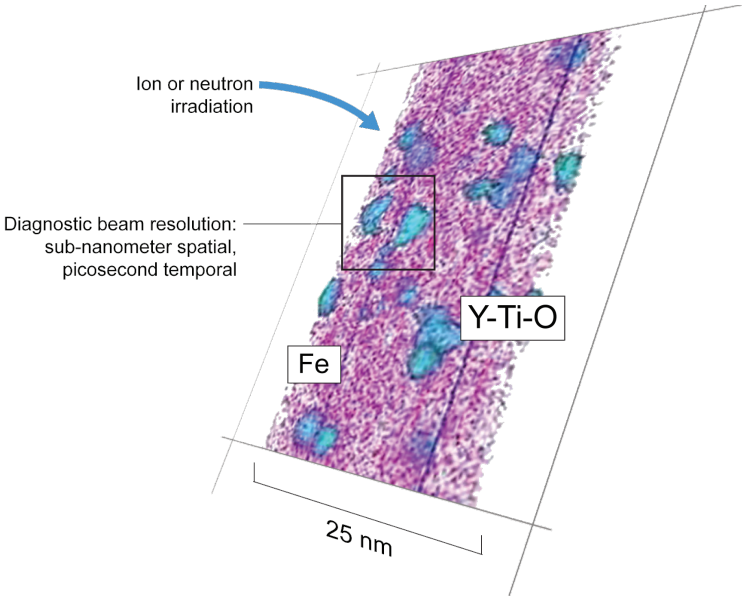
Materials are the primary bottleneck holding back the performance gains and cost reductions of new national security and clean energy technologies.

Materials design and discovery is poised to make a great leap forward by reaching out to modern theory, modeling, and characterization. In situ experiments allow, for the first time, watching structure form during synthesis or fail when exposed to extreme conditions. Computers can now model not only the structure-property relationship of pure single crystals, but also the functionality of multi-phase composite materials that are at the heart of most technologies. Sophisticated synthesis techniques now exist at all scales—nanoscale patterning, self-assembly, thin film deposition, and bulk fabrication. The challenge and the opportunity is to combine these advances in a single giant feedback loop—enabling structure, properties, functionality, performance, and failure to be viewed, understood, and controlled in the same science-based framework. This new paradigm embodies the transition from observation of behavior to understanding and control of functionality and performance. It will accelerate the pace of materials design and discovery to levels needed to support globally competitive national security and clean energy technologies.

The power and reach of this new paradigm is unmatched.

As an example of this approach, MaRIE will provide in situ diagnostic probes, as well as irradiation conditions that mimic those encountered in next-generation reactors, to directly observe these phenomena in nanostructured ferritic alloys. These alloys exhibit enhanced radiation damage tolerance, as well as high strength and microstructural stability over a range of elevated temperatures. These favorable characteristics are attributed to the high density of nanoparticulate oxides distributed throughout the iron matrix, providing interfacial content for recombination and annihilation of radiation-induced point defects, for helium trapping, and for impedance of dislocation motion.

Despite these alloys’ promise in fuel cladding applications, a lack of fundamental understanding of key atomic-level processes encountered during synthesis and service—such as defect-interface interactions and nucleation and growth of nanodispersed oxides in a metallic matrix—limits the development of nanostructured ferritic alloys from widespread application and certification for use in nuclear reactors. MaRIE will enable the development and validation of physics-based, multiscale predictive models, forming the scientific foundation for a process-aware materials performance approach to the design of materials for nuclear reactors.



- MaRIE will guide development of nanostructured ferritic alloys through in situ diagnostics. Shown here is an atom probe micrograph of a nano-ferritic alloy, with iron (Fe) atoms (pink), and other atoms Y-Ti-O (blue).
- MaRIE will provide in situ, time-resolved variants of such data.

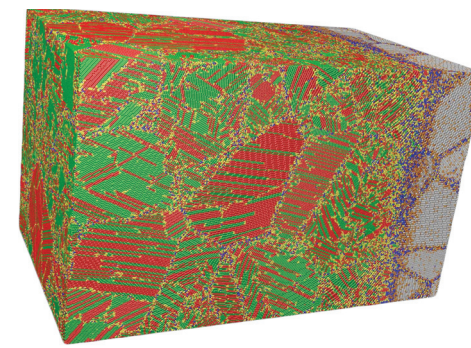
Multi-Probe Diagnostic Hall: Exploring the science of dynamic extremes with MaRIE

The performance of materials in dynamics extremes, also known as compression science, is vital to a broad spectrum of engineering and defense applications related to Los Alamos National Laboratory's national security science mission. In addition to supporting the certification of our nuclear stockpile in the absence of underground testing and a broad spectrum of engineering and defense applications, compression science has altered our view of the material world around us. The discovery of unexpected physical and chemical phenomena and new materials through the application of compression science techniques has led to a new and refined understanding of the nature of chemical bonding in extreme environments. It is clear, however, that many important aspects regarding the response of materials to compressive loading are still not understood, let alone modeled in a predictive mode. As a result, we have not derived the many benefits that a predictive understanding would bring.

To enable the transformation from the present era of observation to that of control of material's functionality and performance, new scientific tools are needed.⁹ MaRIE's MPDH will enable multiple, simultaneous measurements at the micron frontier to address a variety of compelling applications, such as in situ, real-time, three-dimensional, experimental microstructural quantification in extreme environments, multiscale fluid dynamics, or extreme electromagnetic field interactions with matter. Such measurements allow discovery of mechanisms and validation of simulations. MaRIE will provide dynamic observations of microstructure that yield control of materials needed to reduce costs and increase confidence in the nuclear stockpile or advanced nuclear energy systems. In order to meet the facility functional requirements, the MPDH preferred alternative incorporates simultaneous and multiple x-ray, optical, electron, proton, and neutron probes.

⁹ A Summary Report on the 21st Century Needs and Challenges of Compression Science Workshop, Los Alamos National Laboratory, LA-UR-09-07771 (September 2009).

An example of multi-reference x-ray Fourier transform holography (background image) from a tabletop soft x-ray high harmonic source (Opt. Lett. 34, 1618 [2009]).



..... Molecular dynamics simulations are used to model solid-solid phase transitions under dynamic extremes. Connecting such simulations to continuum modeling is a multiscale problem requiring validation.

Five challenges have been identified that capture the scientific needs required to achieve full understanding and that ultimately support our goal of moving from observation to control:

- Acquire time and spatially resolved in situ measurements at all length scales
- Discover new physics and chemistry in extreme environments
- Incorporate material complexity into multiscale simulations to achieve predictive capability
- Unify static and dynamic compression understanding across relevant length and time scales
- Leverage scientific knowledge derived from theory and experiment to the design and control of real materials (i.e., chemistry, microstructure, defects, etc.)

These decadal challenges have been the subject of community discussion, such as the town hall meeting at the 2009 Topical Conference on Shock Compression of Condensed Matter.¹⁰

The scientific functional requirements of MPDH

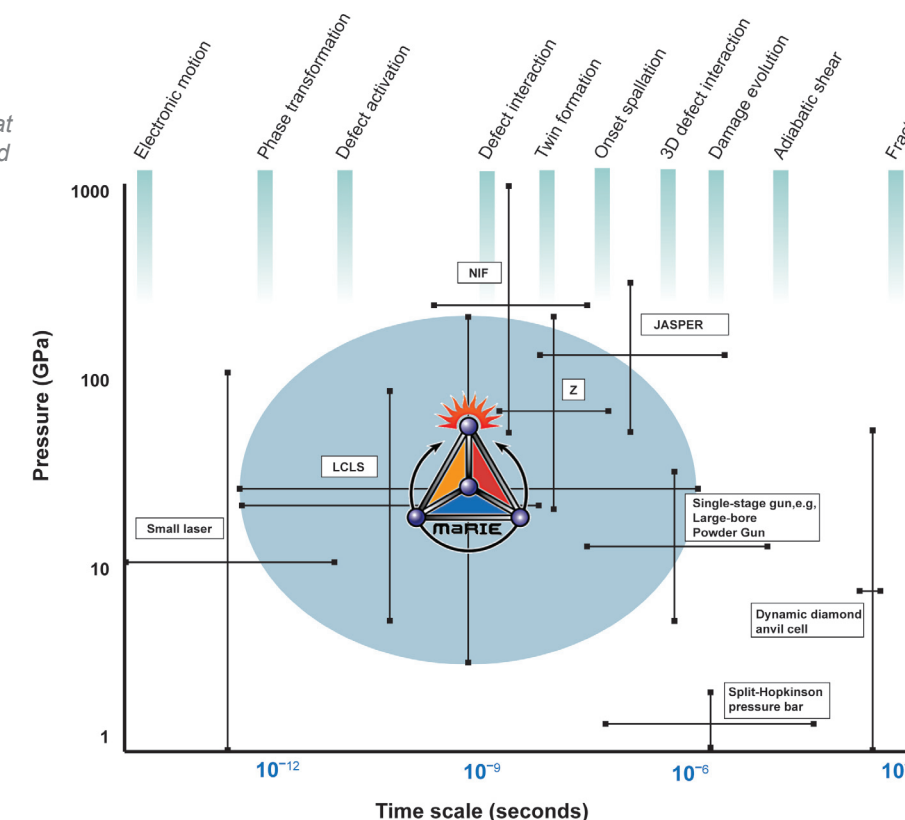
For materials experiments, the relevant length and time scales fall into three clusters: the nanoscale, the mesoscale, and the macroscale. There is strong interest in nanoscale (atomic) dynamics, looking at nanometer resolution on picosecond time scales of phonon motion. This scale is at the forefront of today's science using the current generation of advanced photon and neutron sources. Large-scale integrated experiments at the macroscale will always be needed to confirm the predictions of models. The compelling applications for MaRIE beyond what can be explored using present facilities require understanding the mesoscale of multigranular samples, at the micron frontier.

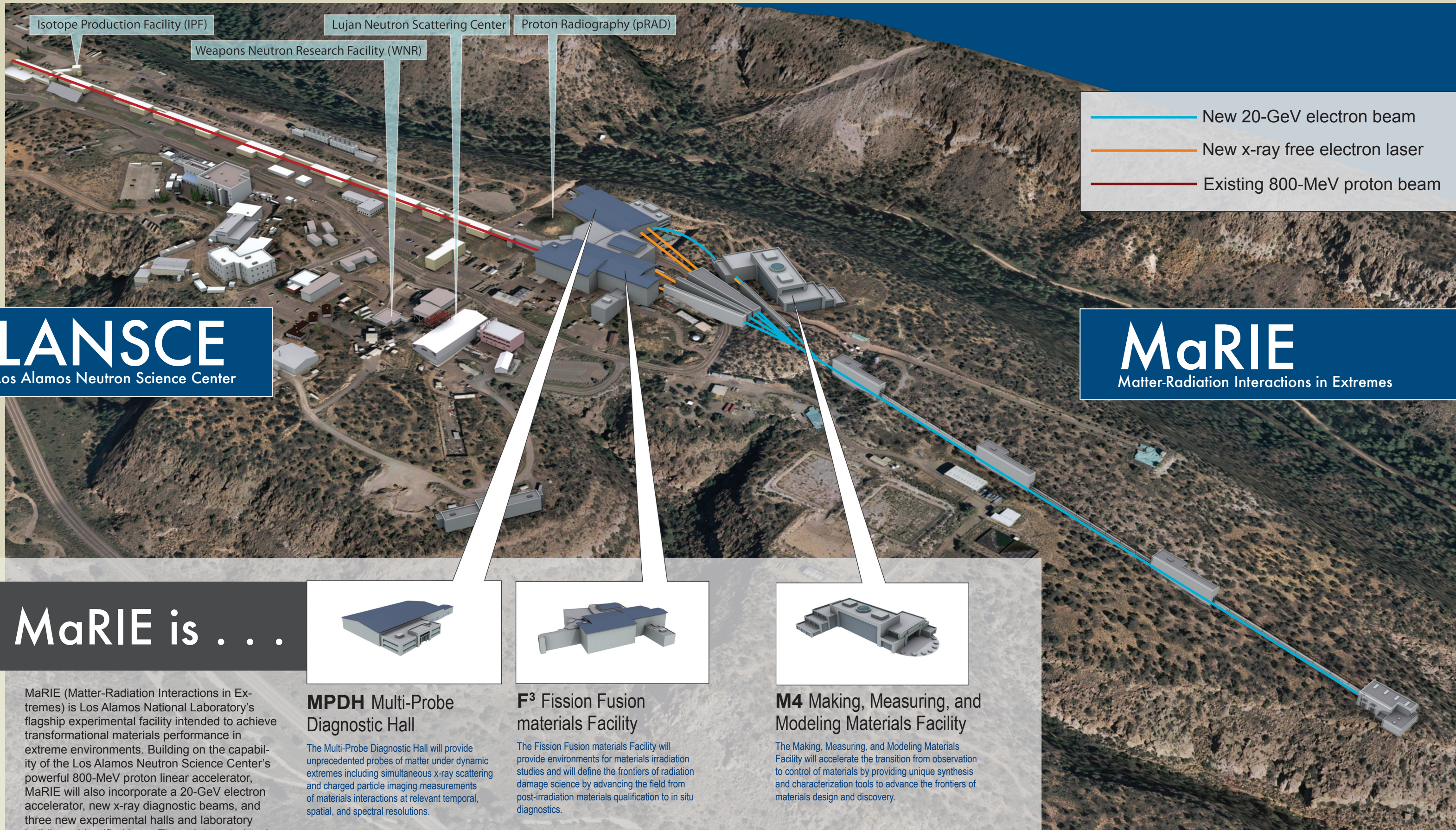
At the mesoscale, it is necessary to be able to create and observe multigranular samples with the properties of bulk material. The initial samples must be synthesized using many different techniques and then carefully characterized; this is a role of MaRIE's M4 pillar. The samples can then be placed into an extreme environment such as high or low temperature or high electromagnetic field. In particular,

¹⁰ David Funk et al., *Proceedings of APS Topical Conference on Shock Compression of Condensed Matter* (Nashville, TN, July, 2009).

Continued on page 20 . . .

..... Mechanisms of material dynamics span many time scales. MaRIE will provide flexible loading capability at key different pressures and rates.



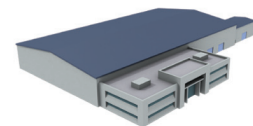


LANSCE
Los Alamos Neutron Science Center

MaRIE
Matter-Radiation Interactions in Extremes

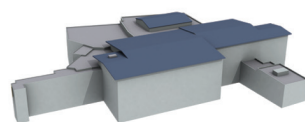
MaRIE is . . .

MaRIE (Matter-Radiation Interactions in Extremes) is Los Alamos National Laboratory's flagship experimental facility intended to achieve transformational materials performance in extreme environments. Building on the capability of the Los Alamos Neutron Science Center's powerful 800-MeV proton linear accelerator, MaRIE will also incorporate a 20-GeV electron accelerator, new x-ray diagnostic beams, and three new experimental halls and laboratory buildings, identified here. These integrated tools will be made available to the external scientific community as an international user facility.



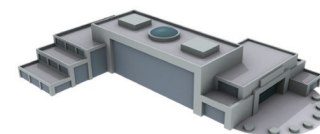
MPDH Multi-Probe Diagnostic Hall

The Multi-Probe Diagnostic Hall will provide unprecedented probes of matter under dynamic extremes including simultaneous x-ray scattering and charged particle imaging measurements of materials interactions at relevant temporal, spatial, and spectral resolutions.



F³ Fission Fusion materials Facility

The Fission Fusion materials Facility will provide environments for materials irradiation studies and will define the frontiers of radiation damage science by advancing the field from post-irradiation materials qualification to in situ diagnostics.



M4 Making, Measuring, and Modeling Materials Facility

The Making, Measuring, and Modeling Materials Facility will accelerate the transition from observation to control of materials by providing unique synthesis and characterization tools to advance the frontiers of materials design and discovery.

. . . the 21st-century facility revolutionizing materials in extremes for transformative materials discovery.

at MPDH, extremes of stress (pressure) over varying strain rates are of great interest; loading of the samples can come from high explosives, flyer plates propelled by various means, magnetic compression, or direct laser illumination. Flexibility in the dynamic extreme environments is critical. The goal of MaRIE is a predictive understanding of dynamic materials performance, not a calibration measurement at a single-point in pressure-density phase-space. As the samples are placed in dynamic extremes, measurements of grain phase, orientation, strain, movement, defect creation and coalescence, and impurity movement are needed, as well as basic density and temperature, to determine quantities such as equation-of-state, strength, and failure thresholds from tension.

The temporal and spatial resolution requirements are set by the need to image and freeze key transient dynamical phenomena in extreme environments. Typical grain sizes in metals and alloys are tens of microns across; spatial resolution of several pixels across a grain region is needed. While the initial defect creation occurs at the nanoscale, the inter-grain void creation occurs on the micron scale. Shock velocities across grains are typically one-to-several microns per nanosecond; thus sub-nanosecond time resolution between separate measurements is needed. The measurements themselves need to freeze phonon motion and phase transformations, thus individual measurements need to be done in less than a picosecond.

Imaging non-periodic features as a function of time in dynamic extremes for national security science is central to MPDH.

As concluded by the Decadal Challenge Workshop of December 2009¹¹, the important science of the future will be driven by rare events, stochastic defects, and in general non-periodic quantities. Hence many measurements require “imaging” or the ability to detect isolated non-periodic structures. Incoherent diffraction from crystal-line structures can provide, at best, two-dimensional distributions of defect sizes. MaRIE will image the inhomogeneities and watch their evolution in time.

Another compelling application for MaRIE is the ability to provide detailed observations of multiscale fluid dynamics. Turbulence of fluid motion is a dominant mechanism in many mission-critical systems; of particular interest is the mixing in variable-density multi-phase flows. Material damage determines the source of some mixing of materials; subsequent turbulent flows are significant contributors to defining the initial conditions for boost and the eventual burn in a nuclear implosion.

¹¹ Decadal Challenges for Predicting and Controlling Materials Performance in Extremes, Los Alamos National Laboratory, LA-UR 10-02959 (April 2010).

The turbulence is driven by large-scale motions, but its dissipation occurs at the smallest spatial scales where viscosity affects tiny eddies of motion (the Kolmogorov scale). The multiple simultaneous probe capability of MaRIE will provide a unique facility capable of observing flows at all relevant scales and drive predictive control of the turbulence.

The high-peak-brilliance XFEL of MaRIE provides 50-keV coherent photons with some pulses available at gigahertz frequencies; this is a unique light source optimized for observations in dynamic extremes. Brilliance refers to the number of photons able to illuminate the experiment in the short time scale of the physics mechanisms. Coherence, or having all the photons in phase with each other, is a property of lasers or small sources of light that allows image information to be reconstructed from measurements of scattered photons. The hard energies are needed to see into and through the required thickness of multigranular materials. Such a light source both penetrates complex, multigranular, mesoscale samples and also does not destroy them.

While the x-ray measurements are important, additional measurements by multiple probes are needed to over-constrain the observations. Capability is needed for simultaneous measurement by multiple probes in order to firmly establish links between microstructure and properties. The inversion algorithm for phase retrieval almost certainly needs “support” (a formal mathematical term as used here) from density information from radiography. The sample’s overall density and size must be monitored in time; temperature measurements in the volume are needed. Charged particle radiography meets many of these requirements, both from the high-momentum electrons of the linac of the light source to provide high time and space resolution, and from protons to provide full sample views.

MaRIE will have several undulators. Multiple “hutches” or experimental areas with an undulator light source available allow multiple teams to perform setup and experiments. Each hutch may have several endstations that share the x-ray beam at different times; each endstation will feature multiple instruments optimized for particular science.

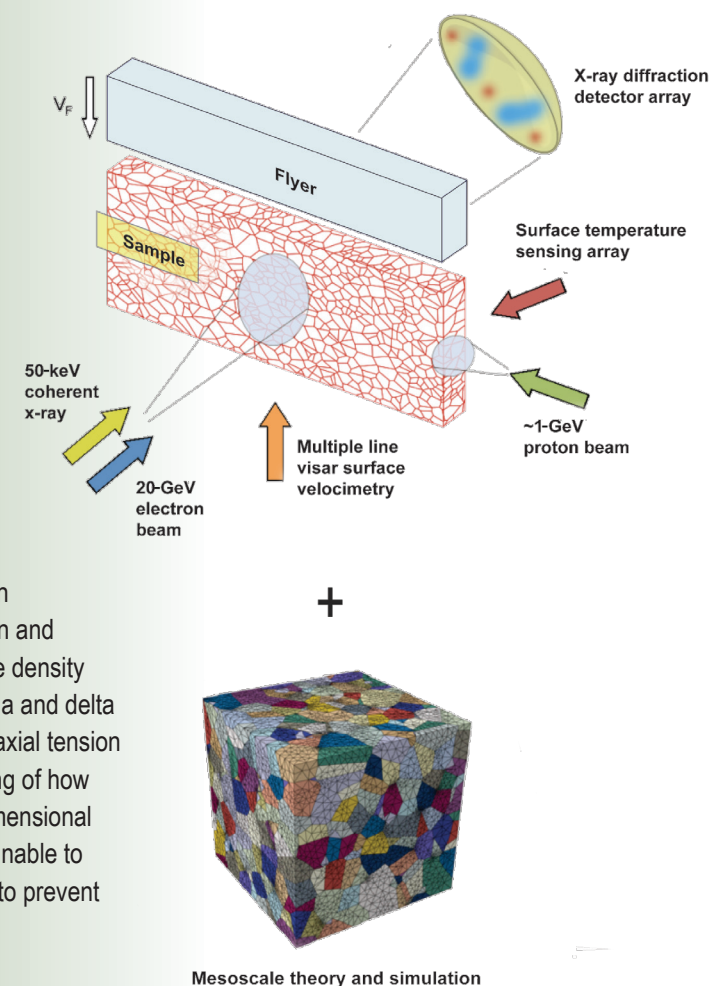
These facility capabilities enable compelling applications as illustrated by descriptions of “first experiments” that would be done for MaRIE. The example described of a multigranular sample undergoing shock loading could be a structural metal, a key earth science mineral, an energetic material (high explosive), or a piece of nuclear material. In all cases it is the unique multi-probe capability of MaRIE that will provide the benefits of predictive capability in compression science.

An MPDH first experiment studying mesoscale material dynamics

The mechanical properties of an individual single crystal depend upon the direction in which it is mechanically loaded. Most metals of strategic and practical interest to our country are polycrystalline aggregates in which multiple crystals and grain boundaries interact under load. Further, most practical metals are either alloys (mixtures of different metals to obtain desired properties) or contain impurities. In either case, there is spatial variability in the chemical composition. This spatial variability in composition will also produce a spatial variability in mechanical properties. There are also other physical phenomena (i.e. twinning, dislocation cells) that occur in polycrystalline metallic materials that produce additional variability. These inherent variations in mechanical properties produce situations where the stress under certain dynamic and shock loading conditions is high enough to tear apart the material.

Under dynamic and shock-driven conditions the loading is many times severe enough to force the material to undergo what is termed solid-to-solid phase transformation or to access other local minima of the free energy in the phase space landscape. Such transitions are between different spatial symmetry arrangements of the atoms comprising the material. These are important since the mechanical properties of each solid phase can be different and the density of each of the solid phases can also differ substantially. Depending upon the material, phase transformation and damage could occur independent from one another or, depending upon the density and volume changes involved, could be interrelated. For example, the alpha and delta phases of plutonium differ in density by 20% and the strain to failure in uniaxial tension is 0.6% for alpha plutonium and 35% for delta plutonium. Our understanding of how these complex dynamic physical events are linked to the detailed three-dimensional nature of these materials is limited. Without this understanding we will be unable to develop predictive models of this behavior or intelligently design materials to prevent damage and failure events under severe loading conditions.

An MPDH first experiment will probe the physics of dynamic solid-solid phase transformation and damage at the length scales at which defects nucleate. This experiment will provide a detailed understanding of these physical processes and the influence that real material composition and structure has on damage events. Making the appropriate measurements for this experiment requires a transverse and longitudinally coherent source of hard x-rays in a single beam. This source will be used to produce x-ray diffraction imaging following the emerging field of ankylography in which a single x-ray beam is passed through the sample and the diffracted image is captured on a spherical x-ray detector. The captured image is used to reconstruct the three-dimensional spatial structure of the sampled material. This technique would be used to probe a shock-loaded sample and produce a three-dimensional mapping of the material as it undergoes deformation, phase transformation and damage. It is envisioned that simultaneous images in time would be collected in order to study the evolution of these complex processes. These experiments would simultaneously be simulated by the appropriate advanced modelling tools to further develop these predictive tools and assist in our interpretation of experimental results.



- Multiple probes performing simultaneous measurements on a shocked sample will elucidate physics at the micron scale at MPDH.

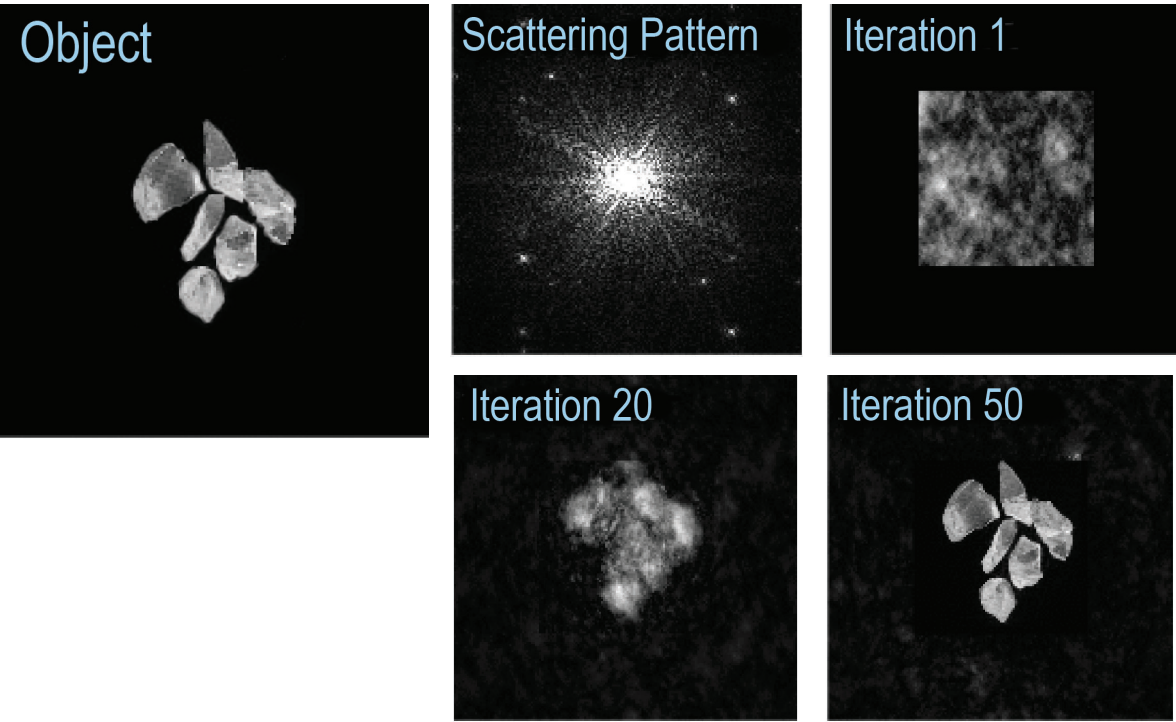
BUILDING MaRIE: Coherent light sources

The need for a coherent light source for MaRIE

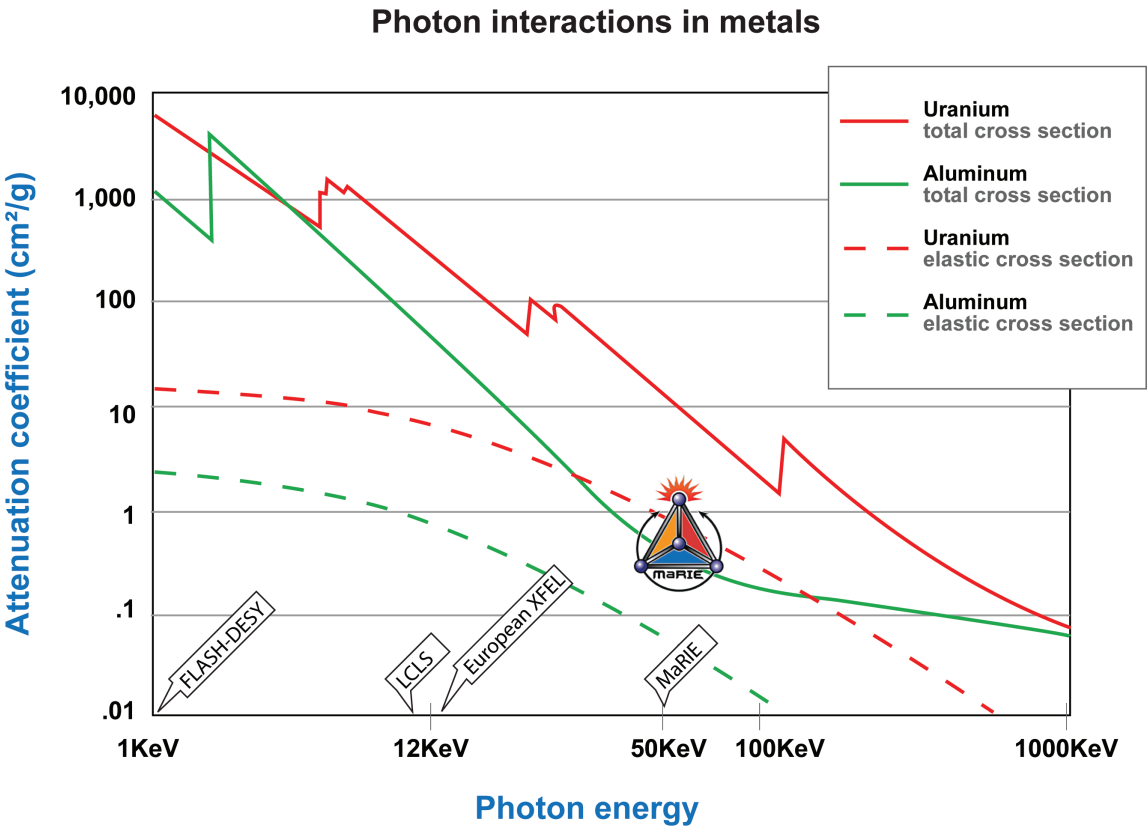
With an infinitely brilliant, totally coherent monoenergetic source of light, three-dimensional imaging of an object using ankylography is possible. Coherently scattered photons from the electron density of an object create a Fourier-transform image in the far-field. However, generally only the real part (the amplitude) of the Fourier transform is measured and the phase information is lost. Using spatially coherent light and “over-sampling” the amplitudes in the image plane can allow the phase information to be retrieved by an iterative algorithm. Today’s state of the art is a thin, flat sample; doing this in three-dimensions rather than for a flat sample requires some longitudinal (temporal) coherence of the x-ray pulse and is much more difficult.

Real experimental limitations result in an imaging figure-of-merit (field-of-view divided by the spatial resolution) that is at best around 1000. In three dimensions, even less spatial resolution is possible. Thus for 100-micron x-ray spot size, perhaps 100-nanometer two-dimensional resolution might be possible and 1-micron resolution in three dimensions is expected.

Higher-energy photons have a lower total absorption cross section and can penetrate thicker samples with more complicated structure and properties representative of bulk material. To see through hundreds of microns to millimeters of metals, energies around and above 50 keV are needed. For coherent diffractive imaging, the useful photons are the ones that are elastically scattered. This cross section also drops with energy, but not as rapidly as the inelastic cross section. By 50–100-keV energies, the ratio of elastic-to-inelastic scattering is peaking, and up to 2% of the incident photons can provide a useful image.



..... An example of two-dimensional coherent x-ray diffractive imaging, accomplished via iterative phase retrieval possible with coherent laser light. Three-dimensional imaging of a sample from a single diffraction pattern is also possible, given a sufficiently brilliant and coherent light source.



..... Penetrating millimeter-thick dense metals requires photon energies of 50-100 keV. Coherent diffractive imaging is optimized when the ratio of elastic-to-inelastic cross sections is large and absorption is relatively small.

MaRIE’s preferred alternative light source, a 50-keV x-ray free-electron laser operating at gigahertz frequencies, allows for unique observations in dynamic extremes.

Too many photons will perturb the sample even to the point of destruction. Present brilliant XFEL sources focus their beams on small (1-micron or so) size spots with few atoms, and the inelastic scattering deposits enough energy to turn the sample into a plasma. The intensity per pulse can be turned down, but then not enough elastic-scattered photons are available to make an image without adding up many pulses. Thus, nanometer and picosecond resolution imaging implies the sample will be destroyed upon each measurement. At these scales, information must be obtained using single photons as with spectroscopy if the sample is to survive or by measuring an ensemble of “identical” samples.

Illuminating a larger spot size and sample volume can reduce the intensity of deposited energy from the x-ray probe, albeit while accepting spatial resolution that is larger. This is exactly the mesoscale on which MaRIE is focused. Electrons in the sample will get an instantaneous increase in energy; how high that can be, how fast the energy drops, and the impact on subsequent measurements is a question of current research. The critical design issue is achieving the proper photon number, taking into consideration the limitations imposed by the source, the sample, and the detector.

In addition to the hard x-ray energy that can allow mesoscale measurements in time scales less than 1 picosecond (faster than the phonon transport time) that do not destroy samples, we also need multiple measurements as a function of time to follow dynamic events. If the electron bunches making the x-rays come from a radio-frequency accelerator operating at a few gigahertz, and if every cycle has an electron bunch, then the x-ray pulses would come a few per nanosecond as needed. Not a lot of pulses are needed at that rate; there are difficulties with beam physics for such closely packed bunches, and maintaining that current would be expensive. Also, detecting so many pulses is a great challenge. An XFEL operating at 50-keV energy with some pulses available at gigahertz frequencies would provide a unique light source optimized for observations in dynamic extremes.

Intermediate steps toward an XFEL

Community-based workshops have helped define an acute need for photons for understanding materials in extreme environments. MaRIE planning has concluded that only an XFEL can meet all the scientific needs, but completion of such a project will take a decade. Thus, we are exploring intermediate steps to generate hard x-ray photons without the full capability of the XFEL we require for MaRIE.

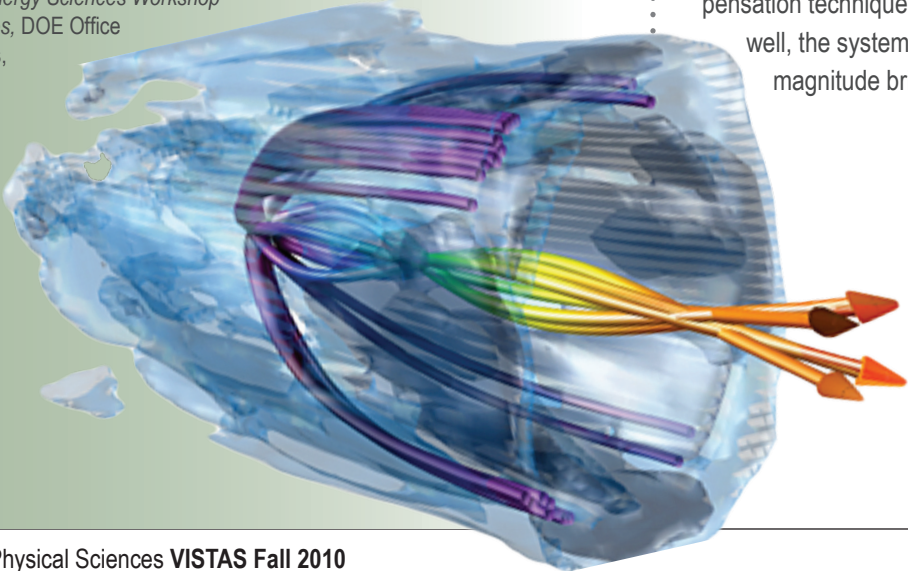
In May 2010 the DOE Office of Basic Energy Sciences convened a workshop on compact light sources.¹² The technology of advanced accelerators is moving rapidly; alternatives for making x-rays for laboratory use are increasing. Inverse Compton scattering sources backscatter an optical laser off an electron bunch and up-shift the energy of the photons. These can create fast pulses of hard x-rays; however, unlike in the XFEL, the light is incoherent and the divergence of the photon beam may reduce the flux inside an extreme environment. Diffraction, radiography, and spectroscopy may be possible with such a source, but not coherent imaging with ultrafast time resolution.

An ultrafast laser pulse scattering off a gas or solid target can create high harmonic generation. With low intensities these can provide extreme ultraviolet photons (less than 100 eV) that are spatially coherent and short (sub-femtosecond) duration. Higher energies may be possible with long-wavelength broad-bandwidth drive lasers. At high intensities with solid targets, x-rays with energies of a few kiloelectron volts can be generated; however, the laser systems must be more powerful and are difficult to run at high repetition rate. Such systems do not penetrate multigranular samples, but can open up ultrafast science.

High-intensity lasers can accelerate electrons to gigaelectron volt (GeV) energies and beyond. Such electron bunches may radiate in the plasma of the target providing broad-spectrum betatron radiation; or they may be injected into magnetic undulators providing XFEL laser light. All these sources tend to be low average brightness, but may be suitable and cost effective for MaRIE endstations as the technical risk is lowered by successful demonstrations of the technology.

¹² Report of the Basic Energy Sciences Workshop on Compact Light Sources, DOE Office of Basic Energy Sciences, Rockville, MD (in press)

• Compact light sources offer a cost-effective, albeit low average brightness, intermediate step to an x-ray free electron laser.



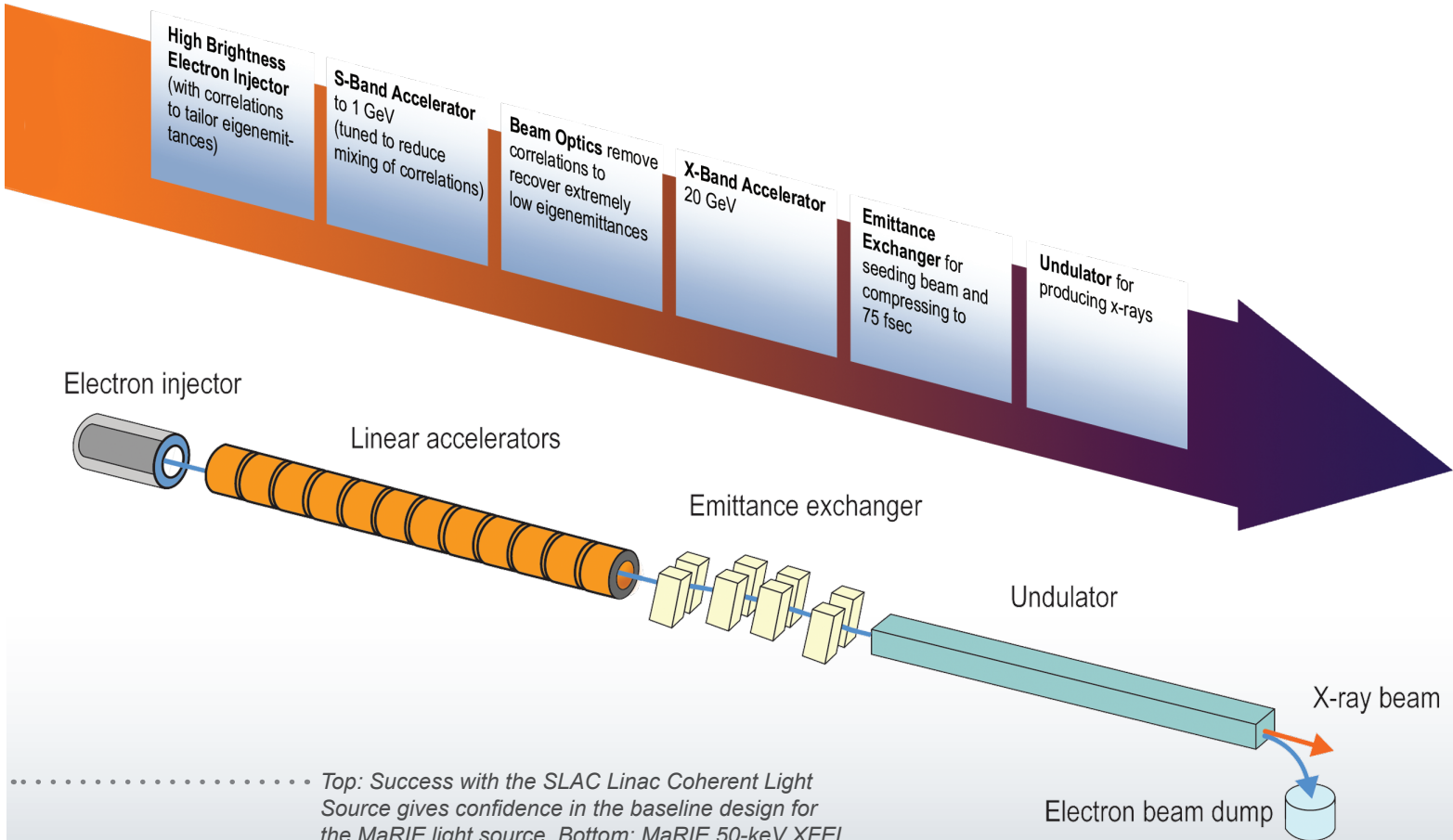
Building the light source required for MaRIE

The preferred alternative for the light source for MaRIE is a very-hard-x-ray (50 keV) high peak, low average brightness XFEL. Free electron lasers are based on technology originally developed at Los Alamos and elsewhere in the 1980s and 1990s. Highly relativistic and energetic pulses of electrons are injected into an undulator, a device with a periodically varying magnetic field. Unlike a usual ring light source where the electrons individually wiggle and emit radiation, in a free electron laser the electrons emit radiation but then coherently interact with those photons and non-linearly bunch up. The bunches then radiate together, causing the total number of photons from a bunch to be proportional to the square of the number of electrons (hence the brilliance) with all the photons being emitted in phase (coherency). One technological challenge is reducing the transverse electron velocity in the pulse—the angular spread in velocity phase space known as the emittance of the beam. For MaRIE, another major challenge will be to reduce the bandwidth of the emission, i.e., to make the photons more monoenergetic, and hence, make the photons also be coherent in time along the pulse.

Current XFEL design capability has become robust with the success of the SLAC Linac Coherent Light Source (LCLS), and accurate estimates of the photon emission can be made. Conservatively using the baseline design with low bunch charge, MaRIE should be able to generate 10^{11} 50-keV photons in a single, less than 100-femtosecond pulse; if emittance exchange or emittance compensation techniques under development work well, the system could be up to an order-of-magnitude brighter.

The total duration of a dynamic event will typically be less than a couple microseconds; such events may be only possible to generate a few times per second. Steady-state turbulent flows require more frequent sampling. Thus, a radio-frequency linac with an accelerating voltage turned on for only 1–4 microseconds, and operated at 10–120 Hertz, can provide sufficient light for dynamic experiments. In radiation extremes at F^3 the thermally and radiation-driven changes occur relatively slowly, and relatively low average brightness is again acceptable.

Building on project success of LCLS and ongoing work on the European XFEL, such a laser has low technical risk—except for the need for longitudinal coherence or very narrow energy bandwidth, which will require development with strategic partners. The electron linac design should be flexible; just as the present LANSCE proton linac has been adapted to a wide variety of uses not originally planned (e.g., proton radiography, isotope production, ultra-cold neutrons), the MaRIE electron linac will become a lasting and valuable resource for great science at Los Alamos.



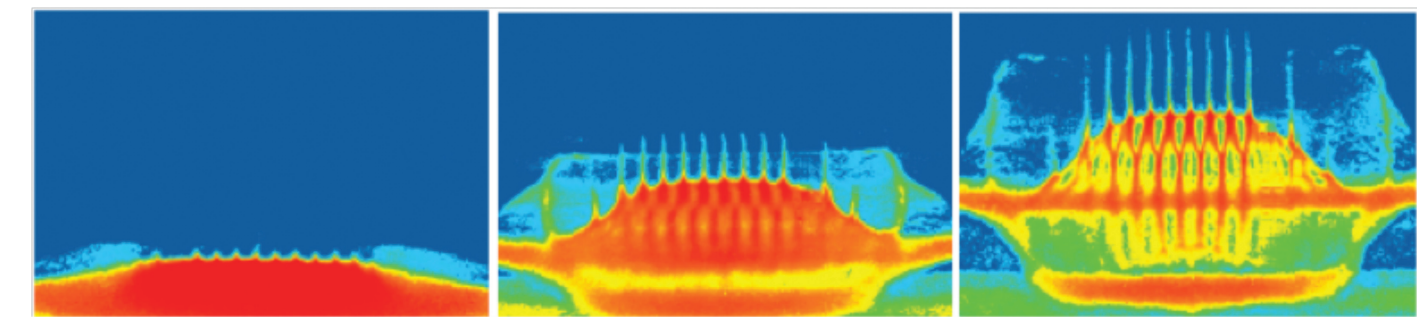
Top: Success with the SLAC Linac Coherent Light Source gives confidence in the baseline design for the MaRIE light source. Bottom: MaRIE 50-keV XFEL baseline concept.

Charged particle radiography

Radiography is the science of generating images of features embedded within an opaque material. Some applications require absolute measurements of the internal density, while other applications require identifying embedded features to study their evolution. Transmission radiography, such as typical medical x-ray imaging, integrates the density along a line through one dimension of a three-dimensional object, providing a two-dimensional image. However, radiographs collected at many angles combined through computed tomography, or assumptions about symmetry of the experiment, can provide detailed information about the density distribution of the three-dimensional object. Multiple radiographs collected at time steps through the evolution of dynamic processes can deliver critical information on the dynamic response to various impulsive loading conditions, such as shock waves or radiation interactions. Los Alamos has pioneered the science and technology of flash radiography since the earliest days of the Laboratory.

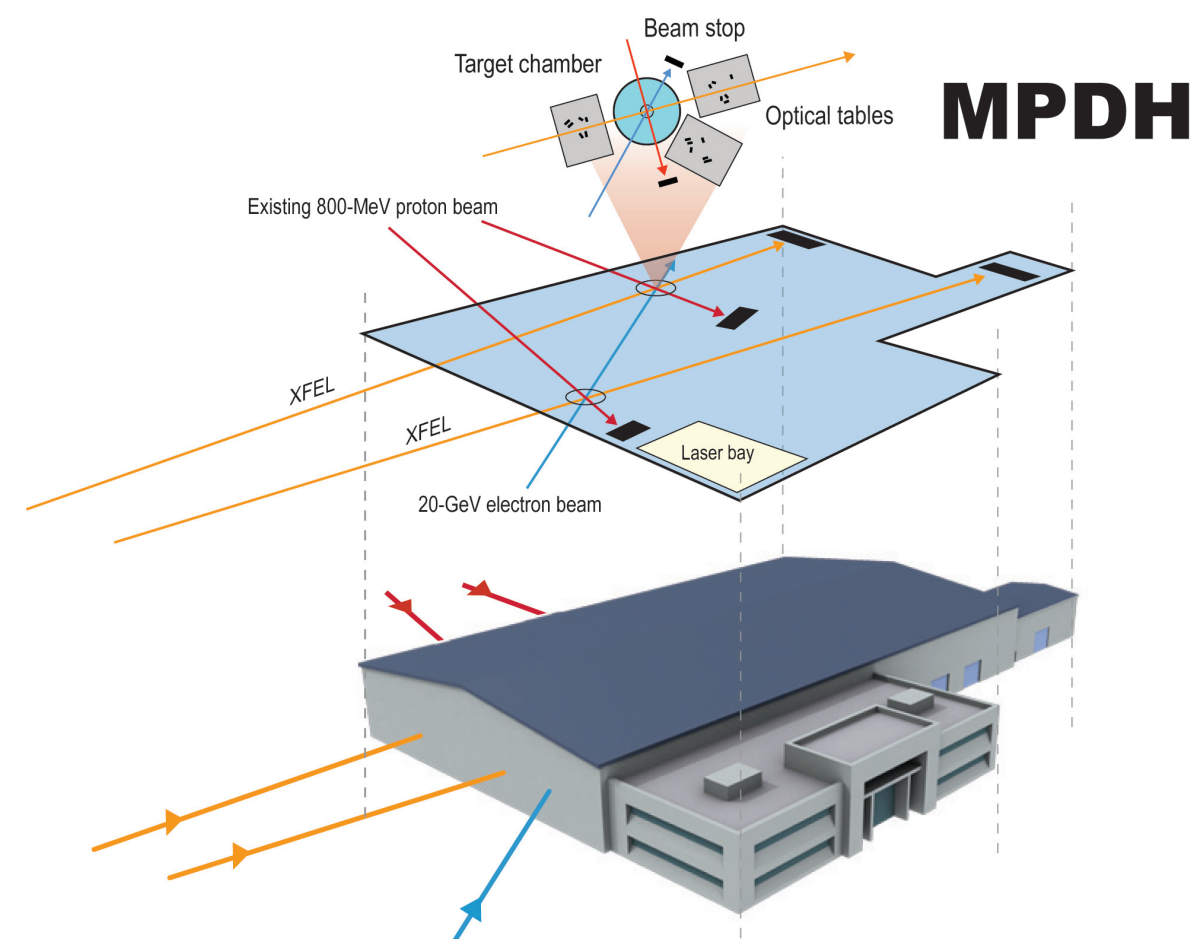
In the late 1990s Los Alamos scientists invented the technique of charged particle radiography, using the 800-MeV proton beam at LANSCE to generate multiple “stop-action” radiographs of dynamic systems during a single event. The intensity of the protons in radiographic images can be measured with scintillators and fast-gated cameras and used to calculate the integrated density through the object. High energy protons are generated with radio-frequency linear accelerator sources, providing a nearly constant stream of particles available to illuminate the sample; observing the radiographs with multiple high-resolution cameras allows the collection of radiographic movies of dynamic events.

MaRIE will exploit LANSCE’s existing 800-MeV proton linac to provide a base radiography capability, enabling 50–100-micron spatial resolution and 50-nanosecond time resolution with 1% density accuracy. This will reveal the temporal evolution of the overall density, shock speeds, volume changes, and sample configuration in dynamic and high-radiation experiments. 20-GeV electrons have nearly the same momentum as 20-GeV protons, and the electron linac for the XFEL can be a useful electron source for high-resolution charged particle radiography complementing 800-MeV proton radiography. In addition to the scattering from the nuclear charge, the electrons radiate photons when they scatter. This “bremsstrahlung” (German for braking radiation) inserts blur into the reconstruction of the scattered electrons, proportional to the thickness of the object being radiographed. However, for heavy metal objects of less than about a millimeter or for thicker samples of lower-Z materials, the resolution degradation is small. Sub-100-nanometer spatial resolution radiographs are possible with the high-energy electrons. A versatile photo-injector for the electron linac can vary the amount of charge for each pulse: small (less than a nano-Coulomb) for x-ray production and large (up to 3 nano-Coulombs) for electron radiography. Thus, multiple high resolution radiographs collected with nanosecond spacing and less than 100 picosecond exposures will allow the study of fast material evolution at a small length scale.

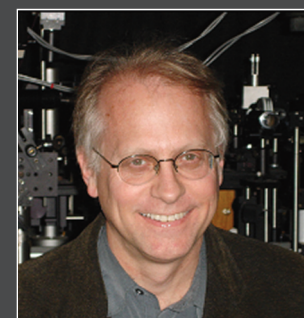


Proton radiographs of ejecta created by Richtmyer-Meshkov instability of shock-loaded surface. Charged particle radiography is ideally suited for generating movies of dynamic events looking through the full samples.

The Multi-Probe Diagnostic Hall will provide multiple endstations where synchronized probe beams are brought together to provide integrated observations of materials in dynamic extremes. X-ray beams (in orange), proton radiography beams (in red), electron radiography beams (in blue), as well as optical laser beams and probes, must be carefully designed to use the available geography.



An expert’s take on compression science



Russell Hemley, Director Geophysical Laboratory Carnegie Institution of Washington

The influence that compression science has had on national security through discovery and application cannot be overstated. In addition to supporting the certification of our nuclear stockpile in the absence of underground testing and a broad spectrum of engineering and defense applications, compression science has altered our view of the material world around us. The discovery of unexpected physical and chemical phenomena and new materials through the application of compression techniques has led to a new and refined understanding of the nature of chemical bonding in extreme environments. However, it is clear that many important aspects regarding the response of materials to compressive loading are still not understood, let alone modeled in a predictive mode. As a result, we have not derived the many benefits that a predictive understanding would bring.

Addressing this challenge will require a new set of characterization tools, temporally and spatially enhanced diagnostics, and the development of new predictive models to characterize the behavior of matter under these extreme conditions. The information gathered in static compression science experiments, when combined with information acquired dynamically, offers the opportunity of understanding the fundamental mechanisms that drive the processes occurring in matter observed throughout the visible universe, ultimately leading to an ability of controlling matter to meet our energy and national security needs.

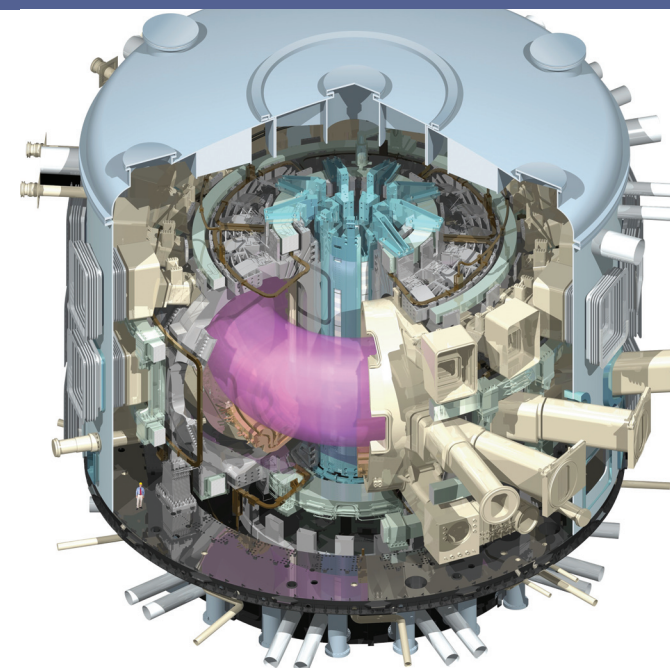
Fission Fusion materials Facility: Exploring the science of radiation extremes with MaRIE

MaRIE's Fission Fusion materials Facility (F³) combines a unique neutron irradiation environment much like that found inside a nuclear reactor with sophisticated in situ diagnostics to "watch damage happen" in materials. This approach to studying materials behavior in a radiation environment will revolutionize our understanding of microstructural physics and significantly accelerate the development of next-generation reactor fuels and structural components.

Nuclear energy is a key element of secure, clean, domestic energy production, and the United States is committed to expanding its reliance on nuclear energy. In this context, the fission community is determining the investment priorities that will extend the lifetime of the current reactor fleet, address new reactors to enable new fuel cycles, develop timely testing, licensing, and certification of new materials, and answer nonproliferation concerns. At the same time, the fusion community recognizes that with imminent ignition at the National Ignition Facility in the United States and construction of ITER, an international magnetic fusion experimental facility, in France, there is a need to turn its attention to the discovery of materials that can withstand the extreme thermal and irradiation conditions produced by a working fusion plasma.

Materials are the immediate priority of both the fission and fusion communities. Extending the lifetime of the current fleet of light water reactors depends on understanding how the materials fail as they age. There are many ideas for a new generation of power reactors that may operate at higher operating temperatures. There is also attraction to using new fuel types that can burn fuel more efficiently, thus extending the time between refuelings of the reactor and extracting more of the energy potential of the fuel. Unfortunately, the time from lab-scale research to certification of a new material for use in a reactor is typically measured in decades. Worse is that the current paradigms for certification require empirical observations in comparable radiation environments. Whereas new fuel cladding and

Nuclear energy (fission reactor core shown in background image) reduces our reliance on foreign oil.



..... A detailed cutaway of the ITER fusion experiment, with the hot plasma, in pink, in the center. ITER is based on the tokamak concept of magnetic confinement, in which the plasma is contained in a doughnut-shaped vacuum vessel.

fuels for existing reactors can, slowly, be certified in the absence of a comparable irradiation environment, there is no proven way to certify new reactor designs without building them and conducting tests—a "catch-22" situation.

The time from lab-scale research to certification of new reactor materials is decades. F³ accelerates the development of nuclear reactor fuels and structural components.

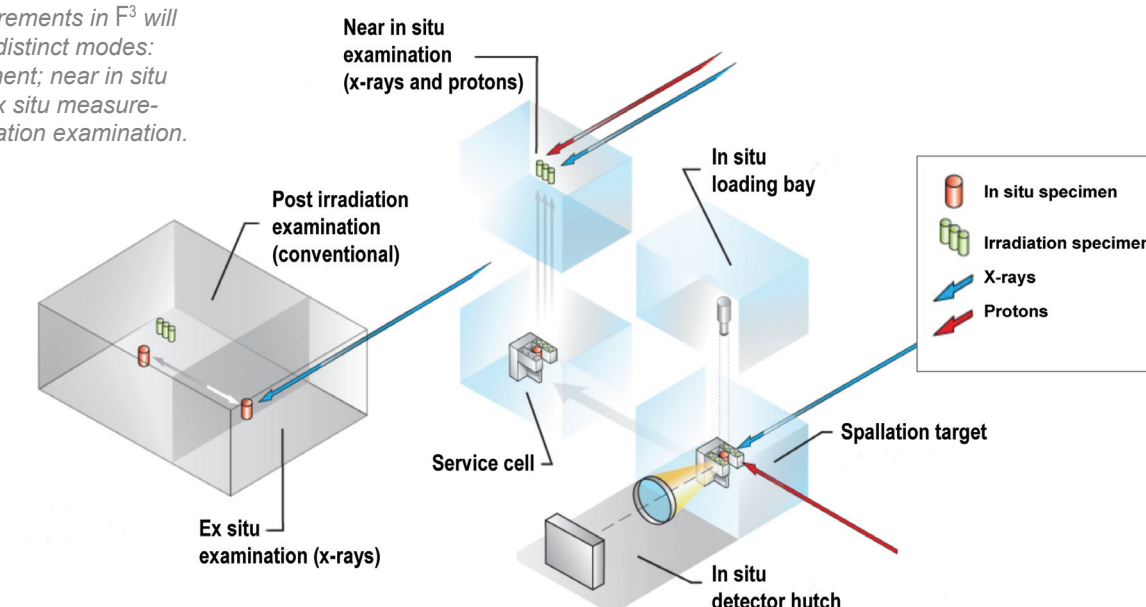
F³ will advance 21st-century radiation damage science research

To revolutionize materials in radiation extremes, MaRIE, through F³, will integrate a spallation-driven neutron irradiation facility with in situ diagnostics of materials using x-ray scattering techniques. At F³, materials data will be collected in a timely and efficient manner over unexplored time and length scales during irradiation on custom specimens in a prototypic radiation extreme environment. MaRIE's irradiation capability will evolve from the Materials Test Station being built at LANSCE for the DOE Office of Nuclear Energy, enhanced with a linac power upgrade to meet irradiation requirements for fusion and other advanced nuclear energy systems. F³ will employ at a suite of locations a set of nondestructive tools based on the unique photon, proton, and electron probes of MaRIE to address four distinct materials characterization needs:

- In situ measurements of custom specimens undergoing irradiation
- Near in situ measurements of specimens undergoing long-term irradiation that are removed temporarily from the irradiation environment for the measurements and reinserted
- Ex situ measurements that are more detailed and extensive than possible near in situ
- Post-irradiation examination on specimens using classical hot cell tools

Some phenomena, like corrosion and creep, warrant direct measurement of material parameters (e.g., temperature and stress) in a radiation environment. Similarly, there are classes of kinetic measurements that occur on short time scales that might be used

..... Materials measurements in F³ will be made in four distinct modes: in situ measurement; near in situ measurement; ex situ measurement; post-irradiation examination.



to validate codes; the short time scales preclude practical measurements outside the neutron environment. In the in situ position it is conceived that samples will be inserted in the radiation field or loaded using a custom carrier if prescriptive environment control is required. Samples will need to be rotated or moved to allow tomography. They will be exposed for as short as a few minutes or as long as several weeks. The high neutron irradiation power on samples creates heating times of a few seconds or less, requiring diagnostics with time resolution under a second.

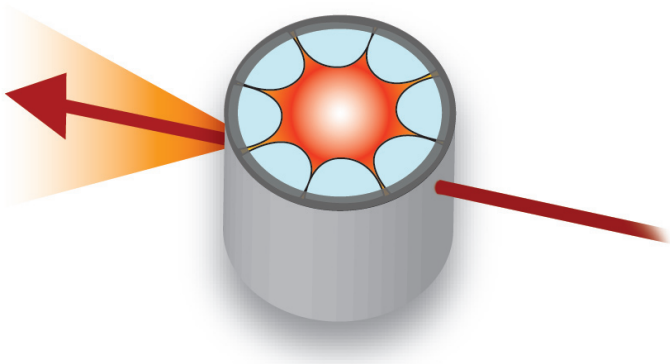
Research on samples undergoing long-term (up to five years) irradiation will benefit from nondestructive evaluation during scheduled beam outages. This will provide researchers extra information about the evolution of sample properties, thus allowing them to act without waiting for the end of the experiment. For example, a new investigation could be started in parallel based on information gained from the present study. MaRIE will allow remote handling of such samples. For these measurements, protons and/or photons could be used.

Instrumentation used for nondestructive examination of samples in the in situ or near in situ locations will likely involve compromises. This argues for an ex situ characterization location optimized for state-of-the-art measurements on radioactive samples, likely in a hot cell remote-handling environment. Properties of the materials resulting from defects that are “frozen-in” after irradiation and cooling can be measured this way.

Sample handling, cutting, and examination will require a suite of tools that are standard in radiation damage and materials science. These are the same requirements used in preparing any samples, except generally with samples that are radioactive. Again, the synergies within MaRIE might well locate the post-irradiation examination (PIE) capability at M4, integrating the radiation extreme environment of F³ with MaRIE’s sophisticated characterization capabilities.

F³ utilizes advanced light source tools in an extreme radiation environment

For many advanced nuclear energy systems, new materials must first be “discovered.” At present, even the best radiation-resistant materials for high dose applications reach their structural limit below 200 displacements per atom (dpa). Fuel burn-up is limited to below 20% because the cladding’s mechanical integrity is reduced by radiation damage and elevated temperature. For fusion applications (and some fast reactor concepts) the challenge is even more severe since helium accumulation resulting from (n, alpha) reactions causes embrittlement.



Light source diagnostic for in situ studies of materials undergoing radiation damage. A nuclear reactor fuel pin is shown in illustration.

Within the fusion community, it is widely recognized that the materials necessary to build a commercial fusion reactor do not exist. To go beyond the status quo of the current nuclear reactor fleet or to enable fusion to realize its potential, we must discover and certify new materials.

Advances in supercomputing, new light source capability, and in situ diagnostics transform materials discovery in F³ for nuclear energy.

The materials irradiation and characterization facilities in the United States need new investment, and a new materials paradigm of accelerated discovery and control over performance must be developed.

The current era holds the promise of this new paradigm because of two new technologies and one transformative idea. First, advances in supercomputing enable fundamental aspects of radiation damage to be simulated using atomistic models in which the initial phenomena of radiation damage, i.e. collision cascades, are computed using various multiscale approaches. Second, external capabilities of third- and fourth-generation light sources, like the Advanced Photon Source and the Linac Coherent Light Source respectively, offer exquisite tools capable of resolving atomic behavior at time and length scales inconceivable 20 years ago. Finally, the idea of bringing such tools to make measurements inside a radiation environment allows the understanding of radiation damage needed to achieve controlled functionality of new materials. The expectation is that models can be developed and validated to a satisfactory level for regulatory agencies. These advances would significantly accelerate materials discovery and certification for use in extreme-radiation-environment applications.

MaRIE SCIENCE: Radiation extremes

An integrated approach to radiation damage studies using neutrons and ions for developing predictive modeling tools

New reactor concepts require materials able to resist radiation damage for up to 80 years. The study of radiation damage in materials is facilitated by subjecting samples to bombardment by energetic ions or neutrons. MaRIE will use both approaches in order to maximize data throughput while retaining damage fidelity.

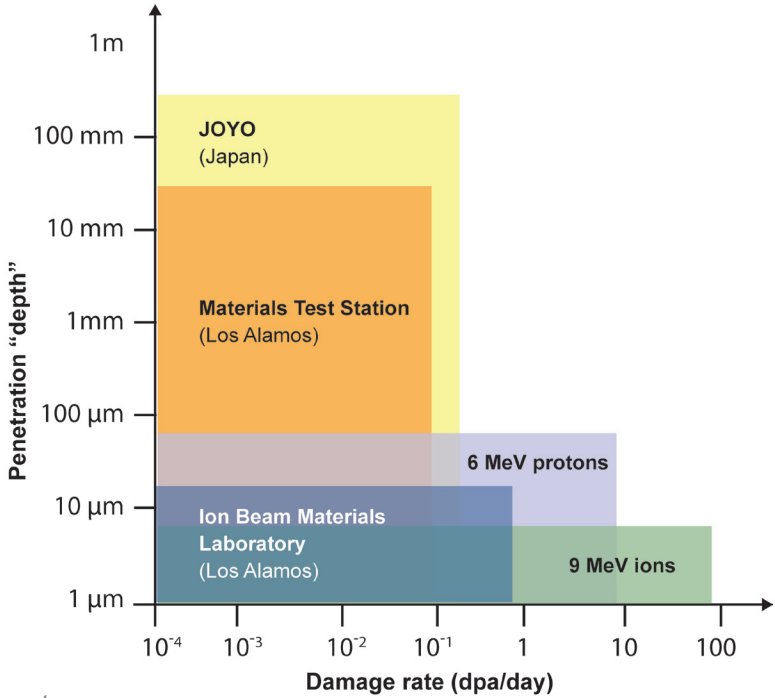
Future advanced nuclear energy systems require radiation-resistant materials that can perform at damage levels exceeding 200 dpa. (1 dpa means that on average each atom in the solid has been displaced from its lattice site once.) The materials experience extreme conditions when high-energy neutrons displace atoms, creating a high density of point defects that then accumulate into displacement damage. This happens at temperatures that can be in the range from 200-1000° C depending on the reactor design, all while in contact with possibly corrosive materials such as fuels and coolants (water, sodium, helium, or possibly lead). Materials under such harsh conditions can experience degradation from mechanisms such as embrittlement, irradiation-induced creep and fatigue, void swelling, fuel-cladding chemical interactions, and stress corrosion cracking.

Qualifying reactor materials and developing new materials for service under high neutron fluxes at the actual operating temperatures is generally performed through testing components or surveillance samples after they have been irradiated in similar extreme conditions. The components or samples are irradiated, removed, and tested by PIE in a hot cell. Facilities performing these irradiations include fast reactors, thermal reactors, and spallation sources, and are limited throughout the world.

Currently, researchers use the BOR-60 (Russia) or JOYO (Japan) fast reactors, producing 30-40 dpa in iron per year; the Advanced Test Reactor (Idaho National Laboratory) and HFIR (Oak Ridge National Laboratory) thermal reactors, which produce 6-10 dpa in iron per year; and SNQ (Switzerland) spallation facility, which can produce 12 dpa per year in iron.

MaRIE integrates neutron and ion approaches to radiation damage investigations for fission and fusion reactors.

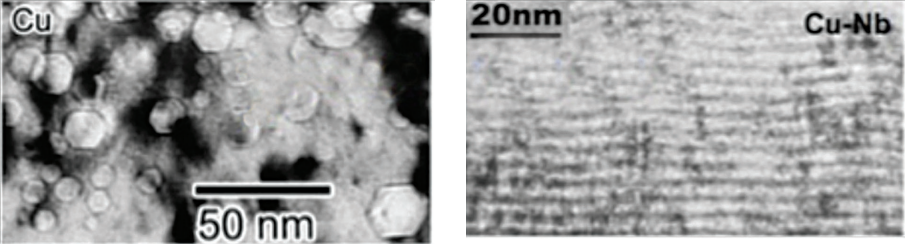
	Advantages	Disadvantages
Protons and Ions	Accelerated dose rate—moderate irradiation times Little to no sample activation Ability to observe damage processes in situ	Small penetration depth
Neutrons	Uniform damage over bulk Damage phenomena are more prototypic of those experienced in fission and fusion reactors	Activated sample typically requiring PIE in hot cells More expensive than ion irradiations Long irradiation times to reach high dose



..... Neutron-induced damage and ion-induced damage are complementary methods for studying materials degradation in a radiation environment.

In conventional neutron irradiation sources, the damage production rate is low, typically less than 20 dpa per year. Thus, it will require 1-2 decades to reach 200 dpa using existing neutron sources. In addition, the surface temperature of the sample and sweep gas chemistry (e.g. fission product gas release) are often the only in situ measurements that can be made.

An alternate means of investigating radiation damage is to use ion irradiation that generates damage approximately 1000 times faster than neutrons. In addition to the high dpa rate, ion irradiation also allows greater control of experimental parameters, including ion energy, dose, and temperature. Ion-irradiated specimens are also generally less radioactive and can be more fully diagnosed in situ. Thus, ion beams provide an invaluable research platform for investigating the unit mechanisms underpinning radiation effects in nuclear energy materials. Both ions and neutrons have advantages and disadvantages for the study of radiation damage in materials. MaRIE, by including both approaches, will provide a unique platform for the development of predictive modeling tools.



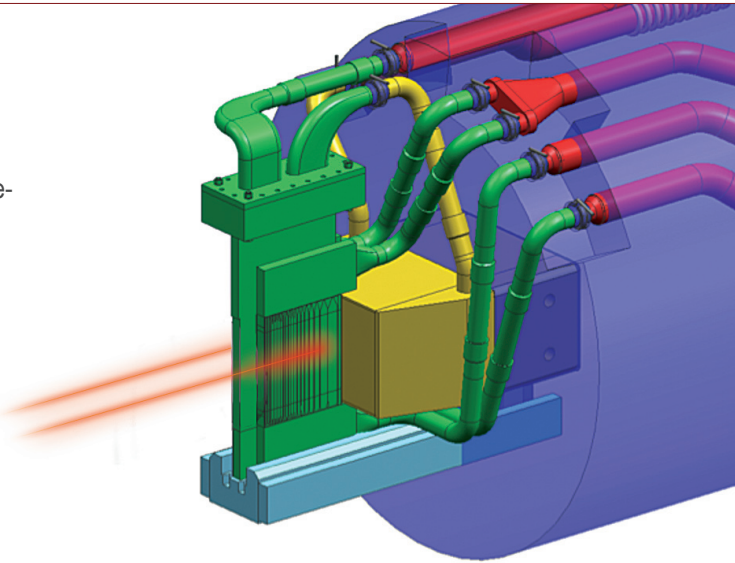
..... Pure copper shows extensive irradiation damage while nanolayered copper-niobium is relatively radiation resistant. MaRIE will enable the study of such radiation damage phenomena with both neutrons and ions.

- Atomistic simulations explore dislocation nucleation at the copper/niobium interface.
- The top crystal is copper and the lower niobium.

The fast neutron irradiation environment in F³

The DOE is developing technologies needed to reduce the quantity of high-level nuclear waste bound for deep geologic disposal. Central to this mission is the development of high burn-up fuel with significant inclusion of plutonium and minor actinides (e.g., neptunium, americium, and curium). The success of this transmutation fuels development program cannot be judged until candidate fuels have been irradiated and tested in a prototypic fast neutron spectrum environment. Unfortunately, only a few fast-spectrum irradiation facilities exist around the world (none in the United States or Europe), and gaining access to them for fuels testing is a complicated, expensive, and time-consuming process.

The Materials Test Station at LANSCE addresses the need for a domestic fast-spectrum irradiation capability. The primary purpose of this facility is to irradiate candidate fuels and cladding in a neutron spectrum similar to a fast reactor spectrum; structural samples will also be irradiated in this same environment. The addition of in situ material diagnostics to the Materials Test Station is central to the design of F³. A close coupling between F³ and controlled materials fabrication in M4, thus leading to predictive models, is at the heart of MaRIE. The basic configuration of the Materials Test Station target system consists of two spallation target regions separated by a fuel irradiation region. The spallation targets are tall, narrow blocks of tungsten cooled by liquid lead–bismuth eutectic (Pb–Bi). The fuel irradiation region is 20-millimeters wide and can hold up to 40 fuel pins, each with an active fuel pellet stack height of 120 millimeters. The pulsed nature of the proton beam allows alternate beam pulses to be directed onto first one, then the other, spallation target in a straightforward and direct manner.



- The proton beam (orange) strikes the Materials Test Station spallation target (green) to produce neutrons for materials irradiation testing.

The reason for using two spallation target sections and alternating the beam pulses between them is to achieve nearly uniform time-averaged neutron flux in the fuel irradiation region, which is located between the two spallation targets. The peak flux in the fuel irradiation region is approximately 2×10^{19} neutrons/ (m²s), a flux comparable to that found in nuclear reactors.

A critical parameter for obtaining meaningful irradiation data is the temperature at which the irradiations are performed. The fuel clad temperature must be controlled within a specified tolerance, and irradiation temperatures up to 1000°C must be attainable in the Materials Test Station. The fuel clad temperature can be controlled by adjusting the Pb–Bi inlet temperature, which can be done in a straightforward manner.

Located laterally outboard of the spallation targets are additional irradiation positions where materials samples may be placed. The neutron flux gradient in the lateral direction is rather high, about 3% per millimeter, yet this is acceptable if the samples are sufficiently thin in one dimension (no more than 250 microns), as is typically the case with modern test specimens. The peak flux in this region is about 20% less than in the flux trap.

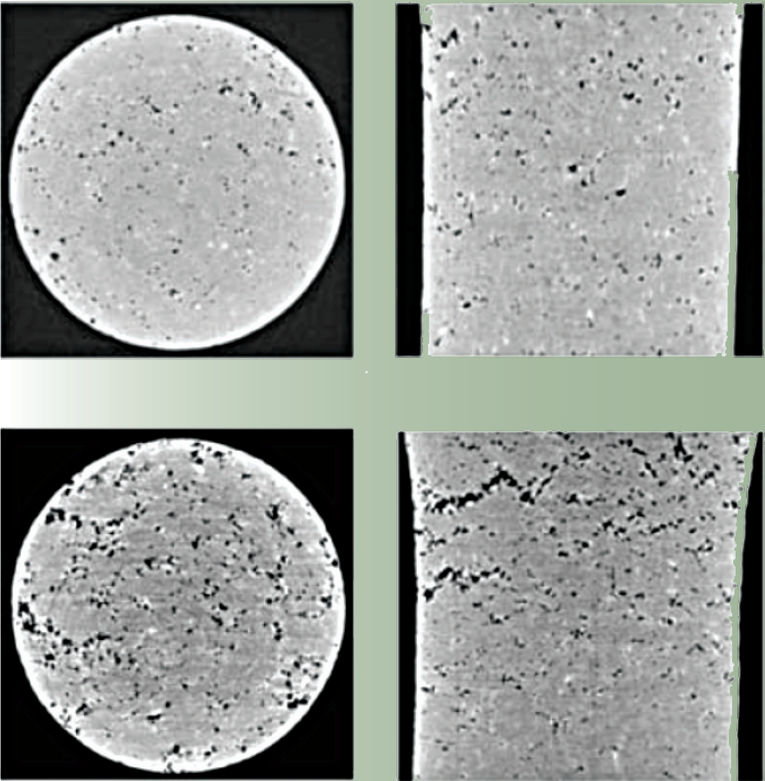
The F³ pillar of MaRIE will differ from all other fast-spectrum irradiation facilities in that it is not a nuclear reactor, and thus facilitates in situ material diagnosis. The primary source of neutrons is spallation reactions resulting from the interaction of 800-MeV protons with tungsten nuclei. The compact design produces a peak neutron flux that is about half of that achieved in the world’s most intense research fast reactors, with the capability to irradiate simultaneously dozens of rodlet-scale fuel pins. In addition to simulating a fast reactor environment, conditions within the Materials Test Station are quite similar in certain respects to those of a fusion reactor, making F³ well suited for conducting fusion reactor materials testing.

Bringing advanced light source diagnostics to F³

Today, bright optical sources provide a probe that can monitor the development of the microstructure of macroscopic solids in real time. Three techniques in particular—high-energy x-ray diffraction, small-angle scattering, and tomography—necessitate instrument geometries that fit well with the diagnostic access envisioned for F³.

The coupling of F³ with a bright x-ray source will provide an unprecedented view of the development of the microstructure (defects, pore growth, cracking, grain development) in situ during irradiation.

Combining advanced light source techniques with the F³ environment will provide a revolutionary leap forward in our understanding of radiation damage mechanisms. With potential technological advances to be made in x-ray beam sources, optics, and detectors over the next decade, new frontiers will open for the study of radiation damage.

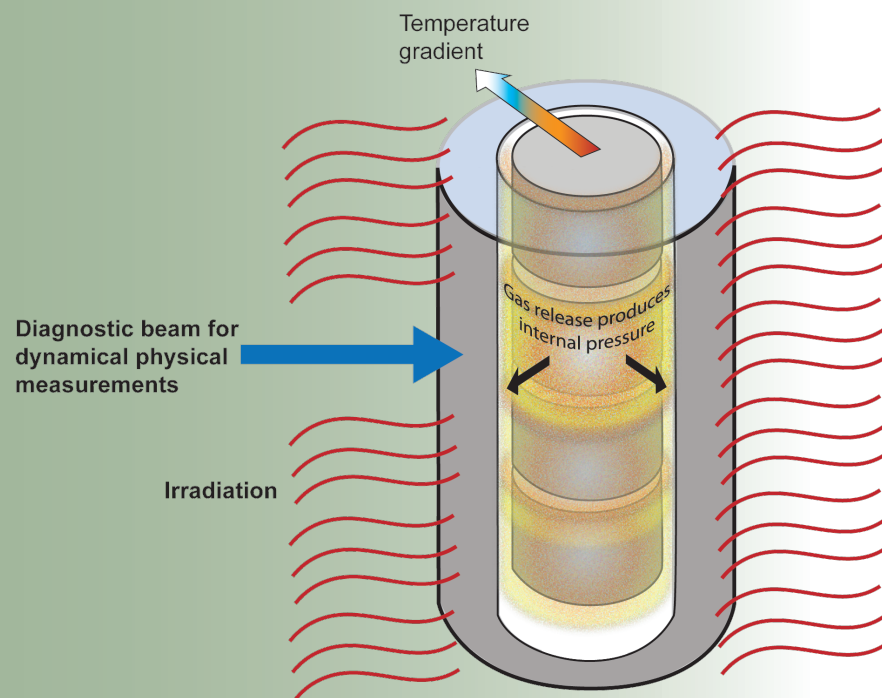


Next-generation advanced light source photon tools will provide unprecedented views of microstructure.

New frontiers of radiation damage science

Design and control of materials able to withstand the extremes of irradiation needed in advanced nuclear energy systems will require experiments that cannot be presently performed. MaRIE will provide capability for critical measurements inside a prototypic reactor irradiation environment, while materials are undergoing radiation damage. Such experiments may include observations of stress and strain, temperature and density variation, and impurity formation and diffusion of transmutation products in fuel pin assemblies. Three-dimensional information can be obtained by tomography when samples are rotated; the time scales required for measurements depend on the phenomena of interest and on the engineering associated with the production of the radiation environment. Accelerator beam characteristics, initial heating during start-up, and sample variations under irradiation are all relevant time scales.

For samples much smaller than an engineering-scale fuel pin MaRIE will enable observations of atomic-scale to mesoscale events for validation of predictive capability of the mechanisms of radiation damage. Thin samples less than 250 microns thick can be subjected to pulsed irradiation, initially from ion sources and later from intense pulsed neutron sources. The dynamic evolution of defects, dislocations, phase transformations, and gas accumulation at interfaces can then be probed in situ.



Engineering-scale reactor fuel pin and cladding prototypes will be irradiated in F³ while diagnostic beams measure physical parameters in situ.

An expert's take

on irradiation extremes



Steven J. Zinkle
Materials Science
and Technology
Division
Oak Ridge
National Laboratory

Imagine a pugilist being knocked to the ground hundreds of times during a boxing match, and each time rising

unfazed with no visible sign of damage and continuing the bout with no degradation in physical performance. This is analogous to the "self-healing" attributes materials scientists are trying to design at the atomic scale into materials to be utilized in future fission and fusion energy systems.

Materials for the structures of next-generation nuclear energy systems must be capable of withstanding intense irradiation fluxes that cause every atom in the material to be dislodged from its original lattice site dozens to hundreds of times over the course of the reactor lifetime. There are five general deleterious manifestations of the displacement damage in materials: radiation embrittlement and loss of strain hardening capacity, radiation-induced or -enhanced solute segregation to grain boundaries and other features which can lead to weakened boundaries or increased susceptibility to localized corrosion, volumetric swelling due to coalescence of displacement vacancies into cavities, radiation induced creep due to preferential bifurcation under applied stress of point defect absorption at dislocations and other lattice features, and high temperature embrittlement of grain boundaries due to the nucleation and growth of helium-containing cavities created from (n, alpha) transmutant helium and displacement vacancies. Using empirical approaches guided by fledgling models and limited experimental data, materials have been developed with adequate resistance to all five of these degradation mechanisms at low doses, or a subset of these phenomena at higher doses.

In order to enable multi-fold enhancement in the utilization of fission and fusion energy systems, a key challenge is to efficiently utilize advanced simulation and modeling in concert with new transformational experimental tools to discover scientific solutions that will mitigate radiation damage degradation in materials up to high damage levels. This would usher in a new generation of advanced high-performance nuclear energy systems that more efficiently utilize the energy in the original fuel, reduce the amount of waste per kilowatt hour, and provide high-thermodynamic efficiency electricity and process heat for a variety of applications.

Realizing the vision of MaRIE

In March 2010, Department of Energy Under Secretaries D'Agostino, Johnson, and Koonin provided the following guidance to Los Alamos National Laboratory (LANL) Director Anastasio:

"In the longer term, we would like to continue to engage you in a discussion of the need for high quality experimental science capabilities at LANL to ensure the intellectual vitality of the Laboratory and ensure its continued ability to support important Department of Energy missions. The Department's focus on materials under extreme conditions relevant to fission, fusion and nuclear weapon systems could provide a focal point for your consideration of the longer term science facility development at LANL. We encourage you to continue the process of engaging the broader community in order to frame the key scientific challenges in this area for the next decade. We look forward to hearing of your continued progress in defining the path forward."

"MaRIE will revolutionize materials in extremes, a key challenge that spans a broad range of DOE missions,

Director Anastasio responded, "We have been studying the issue of long-term science facility development for about five years. We have concluded that MaRIE, for Matter-Radiation Interactions in Extremes, is the appropriate signature experimental facility for LANL. MaRIE will revolutionize materials in extremes, a key challenge that spans a broad range of DOE missions, through unprecedented in situ, transient measurements of real materials in relevant extremes, especially dynamic loading and irradiation extremes."

MaRIE will build on existing DOE investments at LANSCE, in Los Alamos National Laboratory's materials infrastructure (e.g., Materials Science Laboratory and Sigma Complex), and Office of Basic Energy Sciences national user facilities (e.g., Lujan Neutron Scattering Center and CINT). MaRIE also leverages current and planned investments such as the National Nuclear Security Administration's LANSCE Linac Risk Mitigation effort and the Materials Test Station, a fast-fission irradiation capability being developed by the Office of Nuclear Energy. Fully developing the tools that constitute MaRIE will likely take a decade or more; in the meantime, Los Alamos will continue to steward and invest in these foundational capabilities.

The Laboratory has worked aggressively to engage the external scientific community in the development of MaRIE. During 2009, the Laboratory hosted five different workshops culminating in December 2009's "Decadal Challenges for Predicting and Controlling Materials Performance in Extremes," which taken together engaged more than 225 scientists from 80 institutions in identifying scientific challenges and research directions to achieve predictive materials performance in extreme environments, focusing specifically on needed capabilities and tools (please see inside back cover).

Further, the MaRIE team has developed a "First Experiments" process modeled after those employed successfully by the Linac Coherent Light Source and the National Synchrotron Light Source-II that has further engaged 170 scientists from 60 institutions in 10 countries in translating decadal challenges to specific experiments that could be performed by MaRIE or similar facilities.

MaRIE's current focus is translating the science need developed through workshops and first experiments to a specific facility definition by identifying the scientific and facility functional requirements to perform the proposed first experiments and assessing performance gaps that might exist between current and planned capabilities and those required to meet the identified facility functional requirements.

through unprecedented in situ, transient measurements of real materials in . . . dynamic loading and irradiation extremes."

At its most recent meeting in May 2010, MaRIE’s External Advisory Board validated MaRIE’s readiness to make the transition from science need to facility definition. Key next steps include finalizing an acquisition strategy, developing a credible cost estimate, and exploring systems engineering considerations including staged development of MaRIE capabilities.

Realizing MaRIE’s full suite of capabilities requires developing and integrating a suite of resources including i) proton accelerator systems and the use of protons both as a source of spallation-derived neutrons and as a diagnostic tool, ii) an electron accelerator for charged particle radiography and to drive a suite of undulators to produce hard, brilliant, and coherent photons, and iii) synthesis and characterization facilities for materials discovery and design including the direct integration of theory. A systems engineering effort is being developed to coordinate and phase these various elements. For example, research and development is required to reduce risk associated with a very high energy XFEL whereas high power proton systems are presently at a greater state of technical readiness. As part of this phasing effort, less capable sources of photons are being explored to enable early in situ science (e.g., an inverse Compton scattering source for initial in situ radiation damage studies). Finally, a cost range estimate for MaRIE, including a rigorous approach to basis of estimate and associated contingency, is currently being performed and will be externally peer reviewed.

Los Alamos believes that a mission need exists for a facility focused on the challenges associated with materials in extreme environments exploiting in situ transient measurements on real materials in relevant extremes. Further, the Laboratory believes that MaRIE is a strong candidate to be the preferred alternative for meeting that mission need. MaRIE is ready to make the transition from science need articulation to facility definition. To do so requires a deliberate focus on acquisition strategy in partnership with DOE leadership and the development of a preconceptual design. Los Alamos is currently performing a preconceptual design study, including the development of comprehensive scientific and systems requirements, analysis of alternatives that would meet those requirements, trade studies that would assess the cost-risk-benefits of a variety of technical options, and estimates of cost and schedule ranges for the full suite of MaRIE capabilities and their associated annual operation costs.

For more information on MaRIE, please contact the MaRIE Program Office at 505-667-3621 or visit marie.lanl.gov.

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Experimental Physical Sciences mission
We develop and apply a broad set of capabilities in materials science and experimental physics to programs and problems of national importance. The success of our science requires world-class research and processing facilities, including our Los Alamos-based national user facilities—the Los Alamos Neutron Science Center, the Center for Integrated Nanotechnologies, and the National High Magnetic Field Laboratory.

Experimental Physical Sciences vision
We cultivate a responsive, high-performance staff that conducts innovative, cross-disciplinary research and development that produces breakthrough solutions to the most pressing national security challenges.

..... In 2009, Los Alamos National Laboratory hosted five workshops tailored to help identify priority research directions and the capabilities required to meet decadal challenges in predicting and controlling materials performance. The final workshop, “Decadal Challenges for Predicting and Controlling Materials Performance in Extremes,” culminated the series.

The performance of materials in extreme environments is central to a number of national security challenges, including the need for sustainable energy solutions. From fission and fusion energy to nuclear weapons to a broad suite of renewable challenges, a science-based approach to certifying materials performance for extended lifetimes is needed. These workshops, set against a backdrop of urgent mission need and high scientific potential, focused on the means to meet these challenges, emphasizing specifically needed capabilities and tools to seize the opportunity.

The workshop reports are available at www.lanl.gov/source/projects/marie/workshops.shtml.



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U.S. Air Force photo by Master Sgt. Val Gempis
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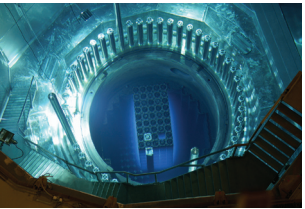


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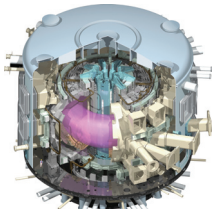


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